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Spectral Transmission Properties (0.35–25 μm) of Some Anthropogenic and Natural Atmospheric Aerosol Constituents

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ABSTRACT

Atmospheric aerosols can have wide-ranging effects on the Earth's atmosphere, hydrosphere, and biosphere. To help improve our understanding of these effects, we have conducted spectral reflectance and transmission properties of a suite of anthropogenic and natural aerosols across a wide wavelength range (0.35–25 microns). Our sample suite included rock and mineral dusts, sulfates and nitrates, ocean water precipitates, and a variety of carbonaceous and organic materials. These data are useful to identify species that may be present in the atmosphere from spectroscopic measurements. Different aerosol/dust species can have unique spectroscopic properties in terms of diagnostic absorption bands and spectral shapes, and diagnostic absorption bands, can be present in multiple wavelength regions, including within atmospheric transmission windows. The composition of aerosol species can be determined with varying degrees of specificity from their spectroscopic properties. Increases in aerosol abundance, which may lead to saturation of strong absorption bands can be partially compensated for by accompanying strengthening of “secondary” absorption bands that are normally weak. Grain size variations mostly affect absorption/transmission intensities, but do not lead to the appearance of any new absorption

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bands. Empirical laboratory studies, where grain size and concentration are varied, can provide information that can be used to determine atmospheric aerosol composition and to help constrain measurements and models of atmospheric optical depth.

Keywords: Atmospheric Dust; Aerosols; Spectroscopy; Transmission; Reflectance

1. Introduction

Dust in the Earth's atmosphere can have wide-ranging and significant impacts. For instance, atmospheric warming or cooling effects due to dust are often uncertain and depend on a number of factors, including optical properties^[1–4], composition^[5,6], and grain size^[7,8]. Dust is both influenced by and influences the Earth's atmosphere, biosphere, and hydrosphere in multiple ways. These effects are normally composition-specific and can include:

- Cloud nucleation^[9–12], cloud reflectivity^[13,14], and precipitation processes^[15–18];
- The sign^[3,19–22] and magnitude of atmospheric radiative forcing^[23–27];
- Hydrologic cycles^[28–32];
- Terrestrial^[33,34] and marine^[35–37] biogeochemical cycles;
- Vegetation health and productivity^[38–41];
- Soil and water fertility^[34,38,42,43];
- Water pollution^[44,45];
- Food production^[46,47]; and
- Air quality^[48,49] and human health^[50–54].

Identifying specific atmospheric dust components can also aid in identifying dust emission sources, as many dust components can be concentrated nearest their source regions^[55–57]. Deserts are an important source of atmospheric dust^[58]. Dust source identification was the primary goal of the Earth Surface Mineral Dust Source Investigation (EMIT) instrument aboard the International Space Station. It mapped reflected light from the Earth's surface across the 380 to 2500 nm range with 7.4 nm spectral sampling^[59,60]. A knowledge of dust optical properties can also inform possible effects of solar geoengineering projects that involve the injection of dust/aerosols into the stratosphere^[61].

To better understand the impacts of dust on the atmosphere and terrestrial ecosystem, knowledge of its composition^[7,56] and associated optical properties, as well as physical properties (such as size and morphology), is essential^[62–66]. As the atmosphere is a dynamic environment, dust composi-

tion and abundance are constantly changing both temporally and spatially^[67]. To fully capture this dynamism^[68,69], information on the nature of dust is most effectively acquired using remote sensing instruments^[70–73]. Combining various observational techniques, data sources, and models can help improve dust quantification^[74]. Linking observational data to atmospheric dust for climate analysis depends critically on knowledge of the optical properties of contributing species, among other factors^[69,75,76].

Atmospheric properties such as aerosol optical depths (AOD) are sensitive to the optical properties (e.g., absorption, scattering) of the dust species contributing to the measurements^[77], along with other properties. The physical and compositional properties of many atmospheric dusts can also vary with time^[78,79] due to age-related changes in composition, physical properties, and climatic conditions^[80–83]. This can further complicate efforts to link a specific species of dust to specific atmospheric measurements.

As an example of the importance of knowing the physicochemical properties of dust, radiative forcing by tropospheric aerosols is an important mechanism in terrestrial climate change^[84]. Scattering and absorbing shortwave and longwave radiation, termed the direct effect, is due to aerosols significantly altering the radiative balance of the Earth^[85,86]. The indirect effect of atmospheric aerosols includes the way they modify the reflective properties of clouds, further altering the radiative energy balance^[87]. Alteration of the reflectance properties of clouds by dust can exert a net cooling influence at the Earth's surface in two ways. First, aerosols act as cloud-condensation nuclei (CCN), causing an increase in the number of droplets present in the cloud and a decrease in the droplet size, thereby increasing the albedo of the cloud significantly^[88]. Secondly, aerosols, by decreasing droplet size, increase the life span of clouds^[89].

Both the direct radiative effect and indirect effects of aerosols cause an average net cooling at the top of the atmosphere^[89–92]. The Intergovernmental Panel on Climate Change (IPCC) provides a comprehensive list of the global climate model estimations of the radiative forcing attributed

to both the direct and the indirect effects^[91], and although these models calculate the global average radiative forcing associated with aerosols with an ever increasing degree of precision, chemically distinctive aerosols exhibit different radiative properties and differing effectiveness as cloud condensation nuclei, and thus will have different effects on the radiative energy budget. Yu et al.^[90] stated that uncertainties associated with the direct forcing of aerosols are about a factor of two to three; however, the uncertainties arising from indirect climate forcing can be large, and therefore much more difficult to quantify. The ability to discriminate different types of atmospheric aerosols would allow for a more accurate calculation of the direct radiative effect and a better understanding of the complex interactions associated with the indirect effects^[86,91–94].

As a result, one of the largest uncertainties in climate modeling is the effect of dust, particularly in terms of the relationship between dust composition and radiative forcing^[95]. The nature of the forcing is intimately related to, among other factors, dust composition. Incorporation of dust into radiative transfer modeling often requires simplifying assumptions about the composition (and hence optical properties) of dust. For example, carbon-rich particles can be generated by various processes, such as industrial manufacturing, vehicle exhaust, and biomass burning, and each of these sources would produce different types of carbon-rich particles. Such particles have generally been assigned to one of two main types: black carbon (BC) and brown carbon (BrC)^[96], each having different assumed optical properties^[78,97]. The diversity of different forms of airborne carbon-bearing particles has recently been reviewed by a number of investigators^[83,98–102], who point to the wide range of sometimes conflicting or overlapping terms and definitions used to categorize them.

Atmospheric chemistry can alter the surfaces of dust grains, changing their spectroscopic properties^[103–105]. Additionally, the spectroscopic properties of complex constituents, such as organic dust grains (e.g., brown carbon), can vary with composition^[106–110] and over time^[111–113].

1.1. Purpose of This Study

We have undertaken an empirical study of the spectral transmission properties of a range of natural and anthropogenic dust constituents over a wide wavelength range in

order to:

- (1) Identify spectral “fingerprints” of different species of dust;
- (2) Identify potentially diagnostic absorption features associated with different concentrations;
- (3) Determine how dust concentration may affect atmospheric transmission and absorption;
- (4) Provide empirical data for deriving optical constants and atmospheric opacity.

This study provides spectroscopic data that could be applied directly to atmospheric transmission models. The data can also be used to quantify or constrain optical constants of specific components, and to model the transmission and absorption properties of mixed dust. Because of the wide wavelength range of this study, optical depths can be determined that are applicable to a wide range of models, which normally cover a few discrete wavelengths or a limited spectral range^[114–117].

The majority of remote sensing-based retrievals or characterization of atmospheric dust are somewhat non-specific in the sense that they generally acquire data at a few wavelengths to broadly characterize “groups” of atmospheric dust and aerosols^[118–122]. This limits their ability to identify individual species and can introduce uncertainties in global models^[7]. By covering a wide wavelength range, we can inform the analysis of data from multiple spacecraft missions that use different wavelengths for atmospheric dust characterization.

Optical properties, such as absorption coefficient, can vary by orders of magnitude over short wavelength intervals^[123], making species identification using limited wavelength coverage uncertain. In addition, as radiative forcing is species-specific, it would be useful to be able to characterize aerosol particulates in terms of both species and abundances.

We used transmission spectroscopy to study the optical properties of different dusts. This allows the laboratory data to be compared to atmospheric data measured using solar occultation, a measurement strategy that has been used in the past to study the vertical profile and dust composition of the Earth’s atmosphere^[124]. We also acquired reflectance spectra of different species in order to better identify diagnostic absorption bands.

Each atmospheric measuring instrument or spacecraft may have unique capabilities in terms of wavelength range,

spectral bands, spectral resolution, and observation frequency. Reviews of instruments, spacecraft, and observations can be found in a number of references^[115,125–127].

1.2. Dust/Aerosol Classification

Dusts/aerosols are typically classified into several broad categories based on their origins and chemical compositions. This method of classification can be somewhat arbitrary, as many atmospheric dusts/aerosols exist as internal mixtures^[128]. For the purpose of this study, we grouped our samples into five main categories: (1) primary anthropogenic aerosols, (2) carbonaceous aerosols, (3) secondary inorganic aerosols, (4) mineral and rock dust aerosols, and (5) sea-salt aerosols/precipitates.

Primary anthropogenic aerosols are aerosols produced by industrial, transportation, and other activities^[129,130]. This group contains a diverse range of materials, and thus has a large range of radiative strengths. Major contributors to this group include transportation-induced particulates, cement dust, coal combustion ash, and other industrial/man-made dusts^[129–131]. Due to increasing strict emission regulations in developed countries, aerosols from these activities are thought to have a low effect on the global climate balance; however, with the increase in industrialization, particularly in Asia, the source of these aerosols may drastically increase from current levels^[132].

Carbonaceous aerosols, produced by fossil fuel combustion and biomass burning, are composed of a complex mixture of different carbonaceous materials^[132]. They can form by natural emissions from vegetation or, more commonly, from combustion. They are generally categorized into two types: black or brown carbon, based on their composition and appearance.

The terms black carbon, referring to the strong absorbent component of these aerosols, and elemental carbon, referring to the more refractory part of carbonaceous aerosols, have been considered to be poorly defined terms^[98,128,133]. Andreae and Gelencser^[132] provide a more in-depth account of these terms in an attempt to remove some of the ambiguity, as well as discuss the role of brown carbon, which can significantly affect the spectral properties of carbonaceous aerosols. Generally speaking, we would expect significant amounts of overlap of the various forms of carbon from different com-

bustion sources, as compositional properties of carbonaceous airborne dust show a continuum of compositions, such as O:C and H:C ratios^[79,103,134]. Due to its strong absorbance of solar radiation across a wide wavelength range, black carbon is considered to have a positive forcing effect^[135–137]; however, Andreae and Gelencser^[132] point out that the uncertainties presented by the inclusion of brown carbon into the current models could greatly affect the forcing of carbonaceous aerosols^[138]. It is widely acknowledged that carbonaceous aerosols are compositionally and structurally diverse^[139–141], and can “age” over time^[132,142,143]. Because of black carbon’s strong absorption across a wide wavelength range, it can have an outsized effect on climate^[66]. Carbonaceous aerosols can also be produced by forest emissions^[144].

Secondary inorganic compounds are composed of sulfates^[26,56,129,130,145] and nitrates (primarily ammonium sulfate and ammonium nitrate) and are the product of atmospheric interactions with SO₂ and NO_x gases^[146–150]. Sulfate aerosols exert a strong negative radiative forcing effect, as well as acting as effective cloud condensation nuclei^[86,129,135,137]. While not currently considered to have the same magnitude of effect on radiative forcing as SO₂, future emission scenarios predict a large increase in NO_x^[84,129], therefore strengthening the negative forcing of nitrate aerosols. Furthermore, ammonium and nitrate have been found to increase the scattering efficiency of sulfate and organic matter aerosols^[151].

Rock and mineral dust is second only to sea salt in terms of total mass emission^[87], with the major sources of this aerosol being derived from North Africa, the Middle East, and China^[128]. Volcanic eruptions can also produce large plumes of mineral dust^[128,129]. As these mineral species involve complex mixtures, regional compositional differences, and a wide array of particle sizes, uncertainties exist in the optical properties and radiative effects of this group^[152–154]. Mineral dust can affect regional weather patterns^[31].

Sea spray is the largest aerosol contributor, in terms of mass emission^[87], and forms the majority of aerosols present above marine environments. Emission to the atmosphere is mostly produced from breaking waves and is therefore highly dependent on surface wind speeds, sea state, and atmospheric stability^[155]. It should be noted that the term “salt” is not necessarily an appropriate naming convention for this aerosol; here, we use the term ocean water precipitate to acknowledge

that our samples were produced from ambient temperature evaporation of seawater samples. While the majority of this sea spray aerosol consists of inorganic salts, a significant proportion is organic materials^[87,156]. This group exhibits a net cooling effect at the top of the atmosphere^[128,129,157,158].

2. Experimental Procedure

2.1. Sample Selection Criteria

We selected dust species on the basis of their perceived abundance and importance, while providing a survey of members of the different classes of atmospheric dust, with preference being given to some of the larger contributors to the global dust load. Furthermore, we view this study as a starting point for comparing laboratory-derived spectra to observational data obtained from satellites, and for deriving optical constants from our laboratory spectra.

When present in the atmosphere, individual dust components may be more readily detectable than from bulk pow-

der measurements, as the individual components will not interact as much through multiple scattering, which leads to measurements of multi-component mixtures being dominated by the more opaque components. To provide insights into complex atmospheric dust mixtures, we have included both single components (e.g., minerals) and more complex materials (e.g., rocks) in our analysis.

2.2. Description of Samples

A suite of 20 samples was chosen to represent the major compositional “groups” that contribute to the global atmospheric dust load, as described above: (1) primarily anthropogenic; (2) carbonaceous; (3) secondary inorganics; (4) minerals; (5) rocks; and (6) sea spray, and are described in **Table 1**. Compositional analyses for some of the samples are presented in **Table 2**. It should be noted that no one study can cover the wide range of anthropogenic and natural dusts. We selected our samples on the basis of their identification and importance as atmospheric dust components.

Table 1. Description of samples used in this study.

Sample ID	Class	Description
C-47994: Coal fly ash	Anthropogenic	From Geological Survey of Canada
FAN-001: Garage exhaust fan	Anthropogenic	Particulates from garage exhaust fan: Colony Square, Winnipeg, Canada
TIR-001: Tire rubber	Anthropogenic	Abraded from Goodyear P196/R15 tire
CAR-001: Car exhaust	Carbonaceous	Tailpipe of late 1970s El Camino
CHA-001: Chimney ash	Carbonaceous	Bottom ash from wood-burning residential fireplace
NIT004: Ammonium nitrate	Secondary inorganic	Synthetic, Anachemia
SPT101: Ammonium sulfate (mascagnite)	Secondary inorganic	The Geysers, Sonoma Co., CA, USA
SPT105: Ammonium sulfate	Secondary inorganic	Synthetic, Anachemia
CRB145: Siderite	Mineral	Governador Valedares, Minas Gerais, Brazil (for grain size study)
ILL101: Illite	Mineral	Rochester, NY, USA
KAO101: Kaolinite	Mineral	Unknown locality
KFE001: Potassium feldspar	Mineral	Churchill, Canada; extracted from granite boulder
QUA101: Quartz	Mineral	Unknown locality, single crystal
SPT178: Gypsum	Mineral	Long Meg Mine, Little Salkeld, Cumberland, England
ATD-001: Arizona Test Dust	Rock/geological mixture	Powder Technology Inc.
LOE-001: Loess	Rock/geological mixture	Loess deposit: Manitoba, Canada
MSH-001: Mt. St. Helens ash	Rock/geological mixture	Ash from pyroclastic flow deposit
RHY001: Rhyolite	Rock/geological mixture	Tucson, AZ, USA
CSS-001	Ocean water precipitate	Precipitate of sea water from Tulum, Mexico
SS-001	Ocean water precipitate	Precipitate of sea water from Victoria harbor, Victoria, BC, Canada

Table 2. Compositions of some of the samples included in this study.

	Anthro	Anthro	Carb. Gp	Secondary
Wt. %	C-479994	FAN-001	CHA001	NIT004
SiO ₂	13.14	86.65	13.14	0.00
TiO ₂	0.52	0.15	0.08	0.00
Al ₂ O ₃	22.46	3.36	2.55	0.07
(Fe ₂ O ₃)*	(4.51)	(2.06)	(0.77)	(0.04)
FeO	1.17	0.29	0.20	0.00

Table 3. Compositions of some of the samples included in this study.

	Anthro	Anthro	Carb. Gp	Secondary
Fe ₂ O ₃	3.21	1.74	0.55	0.04
MnO	0.04	0.04	0.04	0.009
MgO	15.41	0.25	15.41	1.67
CaO	7.97	28.81	43.57	0.05
Na ₂ O	4.54	18.24	0.71	0.00
K ₂ O	1.381	1.276	17.139	0.004
P ₂ O ₅	0.481	0.091	4.715	0.003
Cl	0.00	18.98	0.50	0.118
SO ₃	0.10	0.09	0.44	0.070
TOTAL	99.862	100.021	99.064	2.037
LOI	0.39	29.76	23.87	99.62
<i>ppm</i>				
Sr	1872	275	1872	20
Zr	362	83	93	29
V	109	36	47	<2
Ni	10	<2	<2	<2
Cr	<2	61	<2	<2
Co	<1	<1	<1	<2
Rb	40	25	90	<1

	Rock	Rock	Rock	Rock	Ocean	Ocean
Wt.%	ATD-001	LOE-001	MSH-001	RHY001	CSS-001	SS-001
SiO ₂	78.28	86.65	60.34	75.48	0.09	0.00
TiO ₂	0.35	0.09	0.92	0.17	0.00	0.00
Al ₂ O ₃	9.95	7.50	17.54	12.86	0.03	0.00
(Fe ₂ O ₃)*	(2.65)	(0.76)	(6.79)	(0.30)	(0.10)	(0.34)
FeO	0.31	0.46	2.48	nd	nd	nd
Fe ₂ O ₃	2.31	0.25	3.70	nd	nd	nd
MnO	0.07	0.02	0.11	0.02	0.00	0.00
MgO	1.65	0.25	2.77	0.10	3.74	3.78
CaO	2.77	0.97	5.54	0.22	0.73	0.85
Na ₂ O	1.77	1.85	4.53	0.17	42.21	39.08
K ₂ O	2.642	1.612	1.23	10.085	0.783	0.821
P ₂ O ₅	0.109	0.057	0.19	0.040	0.014	0.019
Cl	0.00	0.00	0.00	0.00	52.03	54.26
SO ₃	0.07	0.06	0.05	0.06	0.15	1.54
TOTAL	100.311	99.819	100.00	99.865	99.877	100.380
LOI	3.64	0.16	0.98	1.13	83.08	34.89
<i>ppm</i>						
Sr	303	275	564	111	167	178
Zr	214	100	161	276	56	58
V	84	54	166	40	18	23
Ni	7	10	5	5	<2	<2
Cr	106	61	45	71	<2	<2
Co	<1	<1	7	<1	<2	<1
Rb	102	40	25	189	30	50

Note: *: All Fe reported as Fe₂O₃. nd: not determined; Total is expressed on a volatile (LOI)-free basis and with all Fe reported as Fe₂O₃. Where both FeO and Fe₂O₃ are reported, FeO was determined by wet chemistry (WC) and Fe₂O₃ was taken as difference between total Fe and FeO. LOI: loss on ignition = % weight loss after heating sample in air for 60–90 min.

Source: Source and Analysis: Mertzman. Analyses by Stan Mertzman at Franklin and Marshall College (see Mertzman, 2000). Analysis done by XRF.

Particle size has been shown to have a strong effect on the radiative forcing properties of atmospheric aerosols, affecting aerosol life span, reflective properties, effectiveness as cloud condensation nuclei^[129], and atmospheric cooling^[23,159]. While there is some inconsistency as to the size bins for classification, it can broadly be approximated by four size classes: nucleation mode (0.003 μm –0.01 μm), Aitken mode (0.01 μm –0.1 μm), accumulation mode (0.1 μm –2.5 μm) and coarse mode (2.5 μm –50 μm)^[145]. Of these size classes, the accumulation mode affects the radiative balance

the most, as they have the longest lifespan, are highly effective scatterers, and are the most common size of cloud condensation nuclei^[129]. For this study, we produced samples that included a mixture of all four size classes. We also explored the effect of grain size for one of our samples. Particle size can affect the ability to identify the presence of weak absorption bands.

2.2.1. Primary Anthropogenic Group Samples

Tire rubber abrasion particles (TIR-001), coal fly ash (C-47994), and particulate matter collected from a parkade exhaust fan (FAN-001) were chosen as representatives of the primary anthropogenic aerosols group. While most studies have focused on fresh exhaust emissions, in terms of traffic-generated aerosols, recent studies have identified non-exhaust particulate matter as a clear contributor to the traffic-generated aerosol load^[160]. While tire abrasion particulate matter may only be a portion of the total aerosol load, worldwide increases in vehicle use may increase the load of this particulate material. Coal fly ash and other industrially-produced aerosols are among the most widely-regulated aerosol species^[129,130], leading to a general emission reduction in most developed countries; however, distinguishing these aerosol species from others may be of vital importance with the growing industrialization of currently less-developed nations. The parking garage intake-fan filter particulate matter was chosen for inclusion as a general “catch-all” for urban primary aerosols.

2.2.2. Carbonaceous Group Samples

Car exhaust particulates (CAR-001) and chimney ash (CHA-001) were chosen for investigation in the carbonaceous aerosol group. As previously stated, most studies have focused on fresh exhaust emissions, in terms of traffic-generated aerosols^[160]; this study, however, looks at the particulate matter, rather than gaseous exhaust, from an older 1970s-era vehicle. Furthermore, we chose these samples in an attempt to distinguish between the particulates produced from vehicle fuel combustion and those of wood combustion.

2.2.3. Secondary Inorganic Aerosols Samples

Ammonium sulfate and ammonium nitrate were used to determine the spectral characteristics of secondary inorganic compounds. While not strictly from anthropogenic sources, the contribution of sulfur emissions to the atmosphere is pre-

dominantly from anthropogenic activities, rather than natural sources^[161,162]. In order to determine if natural and anthropogenic ammonium sulfate were distinguishable from one another, a naturally occurring ammonium sulfate was also included in this study.

2.2.4. Mineral Dust Samples

In this study, geological dust is represented by both minerals and rocks. Pure mineral species in this study include quartz, gypsum, potassium feldspar, kaolinite, and illite. Potassium feldspar was selected as an approximate analogue of the aerosol dust produced in the Sahara desert, as it is a common component^[163–165] and may be an important ice-nucleating species^[166,167].

2.2.5. Rock and Mineral Mixture Dust Samples

Rock and geological dust samples were included in this study to provide insights into naturally occurring multicomponent mixture spectra. Samples included Mineral mixtures/rocks include rhyolite, ash from the 1980 Mount St. Helens eruption, windblown silt, and Arizona Test Dust.

2.2.6. Ocean Water Precipitate Samples

Ocean water samples were obtained from Vancouver Harbor (Vancouver, BC, Canada) and the Caribbean Sea off the coast of Tulum, Mexico, and evaporated at ambient temperatures to obtain precipitates as representatives of dried aerosols of sea spray.

2.3. Sample Preparation

The solid samples were hand-ground in an alumina mortar and pestle and dry sieved to <45 µm for diffuse reflectance spectral measurements. Samples were then further dry-sieved to <20 µm, and for materials that would be unaffected by contact with water, wet sieved to <10 µm. The wet sieving was accomplished by drawing the sample through a 10-µm neoprene sieve (under vacuum pressure) with deionized water. The sample was then captured in a lower chamber by a 1-µm Nuclepore Track-Etch Membrane (from Whatman Nuclepore). Using the smallest achieved particle size, potassium bromide (KBr; optical grade) pressed pellets were created using concentrations of sample (by weight) of: 0.3%, 6%, and 15%; four pure KBr pellets were also produced as standards. The sample + KBr powder (between 0.75 and 1 g total weight) was thoroughly ground in an alumina mortar

and pestle until the sample was visually homogeneous. This grinding also reduced the average grain size to generally $<5\ \mu\text{m}$. Each mixture was placed in a 25 mm evacuable XRF pellet die and held between two optically-worked mirrors at a pressure of $\sim 16\ \text{t}$ for 2 min while under light vacuum draw pressure. The produced pellets were then stored in a sealed container containing an anhydrous desiccant (Drierite) until spectral acquisition. The iron carbonate sample (CRB145) was also spectrally characterized as pellets containing different grain sizes of the mineral to explore its effect on transmission spectra^[7]. These samples were prepared by hand stirring the sample + KBr mixture to achieve visual homogeneity and then pressing it into pellets. Grinding of the sample was not done in order to preserve the grain size of the mineral.

2.4. Spectral Measurements

The $<45\ \mu\text{m}$ powders were spectrally characterized as bulk dry powders for comparison with transmission measurements. Transmission spectra were acquired using sample powder dispersed in KBr because it offers a number of advantages over other measurement techniques^[168]. Spectral measurements were made using an Analytical Spectral Device (ASD) FieldSpec Pro HR spectrometer that measures reflectance from 0.35 to $2.5\ \mu\text{m}$. Reflectance spectra of the samples were measured relative to a 100% Labsphere Spectralon disk. The sample and standard were measured at $i = 30^\circ$ and $e = 0^\circ$. For each measurement, 500 individual spectra were collected and averaged to improve the signal-to-noise ratio (SNR).

Higher wavelength spectra ($1.4\text{--}25\ \mu\text{m}$) were acquired with a Bruker Model Vertex 70 Fourier Transform Infrared (FTIR) spectrometer. It was equipped with an infrared glow-bar light source, an MCT cryo-cooled detector and a KBr beamsplitter. Reflectance spectra were also measured at $i = 30^\circ$ and $e = 0^\circ$ using a SpecAc Monolayer Grazing Angle Specular Reflectance Accessory. The sample spectra were measured relative to a 100% Labsphere gold standard. 1500 scans of the sample and standard were collected at a scanner velocity of 40 kHz; they were averaged to improve the SNR. This study focuses on transmission measurements; the bulk powder spectra serve as a check on the quality of the transmission spectra.

Transmission spectra were also collected using the ASD

FieldSpec Pro HR spectrometer, and we used a pure KBr pressed pellet as the transmission standard. For the transmission spectra, 100–500 scans were collected and averaged to improve SNR. Transmission spectra in the 1.4 to $25\ \mu\text{m}$ region were collected using the Bruker Model Vertex 70 FTIR spectrometer as described above. Either the MCT cryo-cooled or an RT-DLaTGS detector was used in the acquisition. Transmission spectra were acquired relative to a pressed KBr pellet situated perpendicular to the incident light beam. A total of 100 scans, collected at a scanner velocity of 40 kHz, were averaged to improve SNR.

For this study, atmospheric transmission windows are defined as the areas between: $0.4\text{--}2.5$, $3\text{--}5$ and $8\text{--}13\ \mu\text{m}$, with a few small “dead-zones” present in the $0.4\text{--}2.5\ \mu\text{m}$ window, particularly near the strongest atmospheric water absorption bands near 1.4 and $1.9\ \mu\text{m}$. We focus on fine-grained dust ($<5\ \mu\text{m}$) but also examine coarser dust as size can affect radiative properties^[159], and coarser dust particles may be more important than previously thought^[23]. The reflectance and transmission spectra are provided in the **Supplementary Materials**.

No one technique is perfect in all respects for transmission spectroscopy^[169,170]. Potential limitations include, inhomogeneous pellets, back reflection, and internal scattering.

2.5. X-Ray Fluorescence (XRF) and Wet Chemistry (WC)

Some samples were characterized using X-ray fluorescence (XRF). The analytical procedure is described in Govindaraju, Mertzman, Reichen and Fahey^[171–173], and the website: <https://www.fandm.edu/earth-environment/laboratory-facilities>. Compositional results are provided in **Table 2**.

2.6. X-Ray Diffractometry (XRD)

Some samples were additionally characterized by X-ray diffraction (using a Bruker D8 Advance with a DaVinci automated powder diffractometer or Terra portable XRD. We acquired continuous scan data from 5 to $80^\circ\ 2\theta$; a scintillation counter collected data in increments of 0.02° with an integration time of $1\ \text{s/step}$. The samples were illuminated with Co X-rays, at $40\ \text{kV}$ and $40\ \text{mA}$, and a take-off angle of 6° . Terra portable XRD acquisition involved collecting X-

ray diffractograms using an InXitu Terra 299 portable X-ray device, providing continuous scans from 5 to 55° 2 θ in steps of 0.05°. The Cu source X-ray tube operated at 30 kV and

10 W. Phases identified by XRD are listed in **Table 3**. The X-ray diffractograms are provided in the **Supplementary Materials**.

Table 4. Phases identified in the samples by X-ray diffractometry.

Sample ID	Class	XRD Instrument	XRD
C-47994: Coal fly ash	Anthropogenic	Terra	Amorphous material, quartz
FAN-001: Garage exhaust fan	Anthropogenic	Terra	Dolomite, calcite, quartz, albite, halite
TIR-001: Tire rubber	Anthropogenic	D8	Amorphous
ATD-001: Arizona Test Dust	Rock/geological mixture	Terra	Quartz, albite, microcline
CAR-001: Car exhaust	Carbonaceous	D8	Hematite, quartz
CHA-001: Chimney ash	Carbonaceous	D8	Dolomite, calcite, quartz, halite
NIT004: Ammonium nitrate	Secondary inorganic	D8	Ammonium nitrate
SPT101: Ammonium sulfate (mascagnite)	Secondary inorganic	Terra	Mascagnite
SPT105: Ammonium sulfate	Secondary inorganic	D8	Mascagnite
CRB145: Siderite	Mineral	D8	Siderite

2.7. Expected or Possible Spectral Properties

The samples in the various groups may possess unique or diagnostic spectral properties. Here we summarize possible spectral signatures for the various groups, focusing on the major atmospheric windows: 0.35–2.5, 3–5, and 8–13 μm .

2.7.1. Spectral Properties of Primary Anthropogenic Group

Samples in this group are expected to be dominated by both organic and inorganic (mineral) species. The organic components are expected to exhibit major absorption bands in the 1.7 and 2.3 μm regions from overtones and combinations of the major aliphatic C-H absorption bands that occur in the 3.3–3.5 μm region, as well as 5.5–9 μm due to various C-O fundamentals^[174,175]. Expected minerals, in the coal fly ash, include quartz and feldspars, which are only weakly-featured below 2.5 μm (e.g., AlOH and SiOH absorptions in the 2.1–2.2 μm region)^[176,177]. Trace amounts of iron may cause a reflectance/transmission decrease toward the ultraviolet region and a possible Fe³⁺-O absorption in the 0.9 μm region^[178]. At longer wavelengths, aluminosilicate minerals will exhibit Si-O and Al-O stretching vibrations in the 8–12 μm region.

2.7.2. Spectral Properties of Carbonaceous Group

As with the primary anthropogenic group, possible absorption bands are associated with both organic and inorganic

species depending on the particular sample —see above for details.

2.7.3. Spectral Properties of Secondary Inorganic Aerosols

Ammonium sulfate and ammonium nitrate were used to determine the spectral characteristics of secondary inorganic compounds. Spectra of these materials are expected to be dominated by ammonium absorption bands of increasing strength toward longer wavelengths: 1.05, 1.3, 1.56, 2.02, 2.12 μm (combination and overtones), and 3.05–3.3 μm ^[179]. Absorption bands associated with the sulfate molecule are expected between 8 and 10.4 μm , whose positions are species-dependent; shorter wavelength combination and overtone bands will generally be weak^[180,181]. Ammonium nitrate has N-O absorption features near 9.3 and 12.2 μm ^[182].

2.7.4. Spectral Properties of Mineral Dust

In this study, geological dust is represented by both minerals and rocks. Pure mineral species in this study include quartz, gypsum, potassium feldspar, kaolinite, and illite. Potassium feldspar was selected as an approximate analogue of the aerosol dust produced in the Sahara desert, as it is a common component^[163–165] and may be an important ice-nucleating species^[166,167]. The expected spectral properties of quartz, and potassium feldspar were discussed above. Kaolinite and illite will exhibit metal-OH absorption bands in the 2.2–2.3 μm region, and Si-O and Al-O stretching bands over the 8–13 μm region that are species-specific^[176,183].

2.7.5. Spectral Properties of Rock and Mineral Mixture Dust

Rock and geological dust samples were included in this study to provide insights into naturally occurring multi-component mixture spectra. Samples included Mineral mixtures/rocks include rhyolite, ash from the 1980 Mount St. Helens eruption, windblown silt, and Arizona Test Dust. Their determinable minerals (Table 3) have been discussed above.

2.7.6. Spectral Properties of Ocean Water Precipitates

Ocean water samples were obtained from Vancouver Harbor (Vancouver, BC, Canada) and the Caribbean Sea off the coast of Tulum, Mexico and evaporated at ambient temperatures to obtain precipitates as representatives of dried aerosols of sea spray. Halite itself is spectrally featureless, and any absorption features are due to accessory components.

3. Results

The 20 samples used in this study are presented within the six sample type groupings described above. By measuring transmission spectra of dust in three different concentrations, we are able to identify potentially diagnostic absorption bands for the different materials that may be weak at low concentrations, but could be used to identify the presence of specific species in high concentrations. Different concentrations also allow us to determine approximate atmospheric concentrations where diagnostic absorption bands become

optically saturated and which could negatively impact the derivation of optical constants.

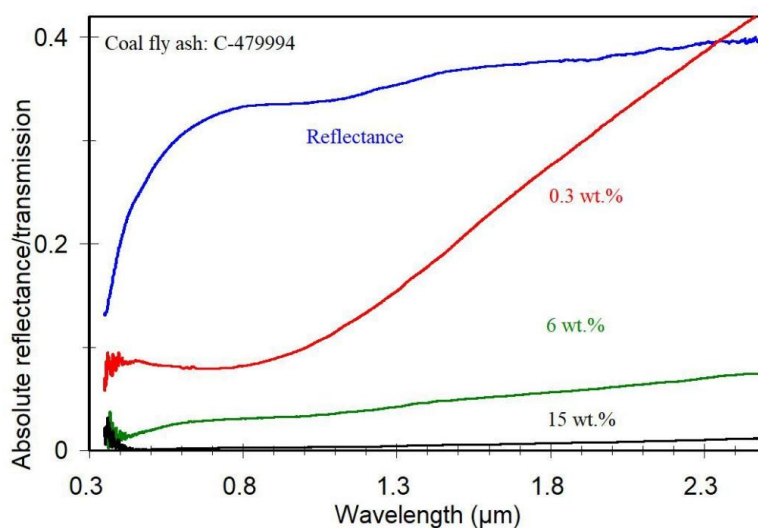
3.1. Anthropogenic Dust Spectra

This group includes coal fly ash (sample C-47994), exhaust fan deposits (FAN-001), and tire rubber (TIR-001).

3.1.1. Coal Fly Ash (C-47994) Spectra

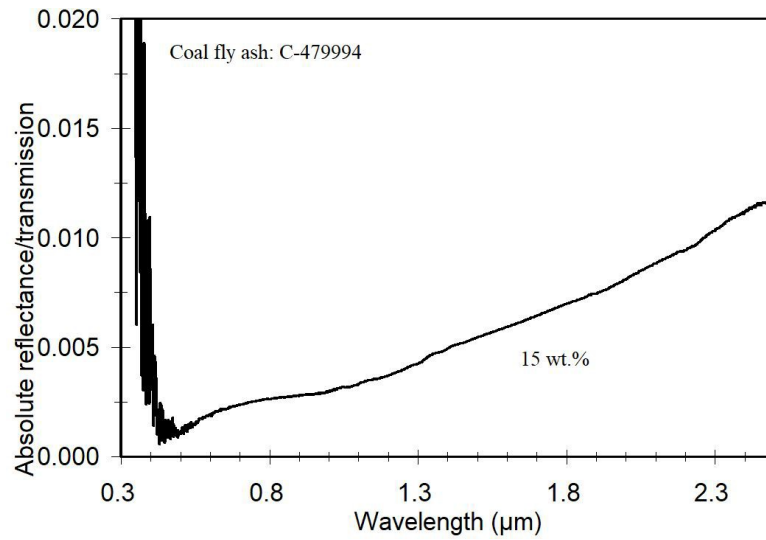
X-ray diffraction analysis showed that the sample is mostly amorphous with a small identifiable quartz component. XRF analysis (Table 2) shows the presence of a substantial aluminum component, which is likely present in the form of amorphous or poorly crystalline aluminosilicates.

The coal fly ash sample used in this study lacks any truly identifiable absorption features in the 0.4–2.5 μm and 3–5 μm transmission windows (Figure 1a,b); a weak but non-diagnostic broad feature exists at $\sim 1 \mu\text{m}$, likely from the presence of Fe-bearing phases in the sample (Table 2). This absorption feature is only indicative of an Fe-bearing component and probably cannot be used to identify specific Fe-bearing phases. Transmission in the visible region decreases slightly from 0.45 to 0.75 μm (from 8.7 to 8.0%) for the 0.3 wt.% sample spectrum. At the higher concentrations, the spectra become more red-sloped (i.e., transmission increasing toward longer wavelength), and are also less transmissive overall (5%) in the visible region. No appreciable absorption features appear, even at the highest concentration (15 wt.%; Figure 1b).

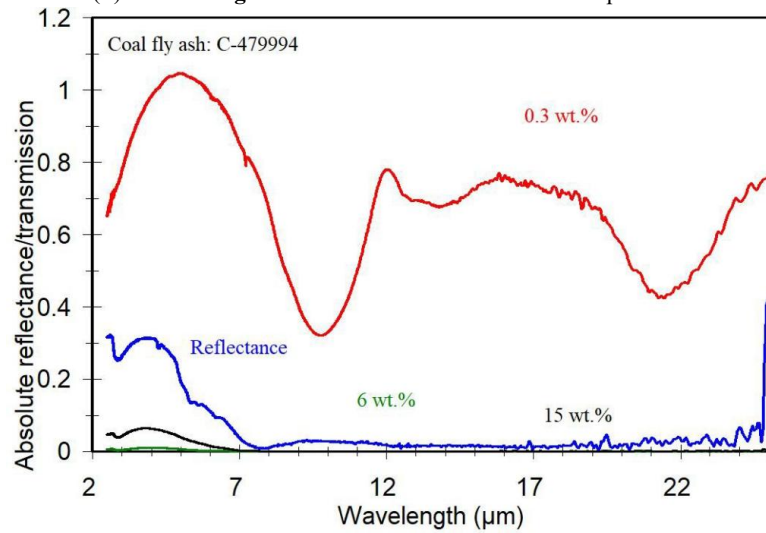


(a) 0.35–2.5 μm reflectance and transmission spectra of coal fly ash sample C-47994.

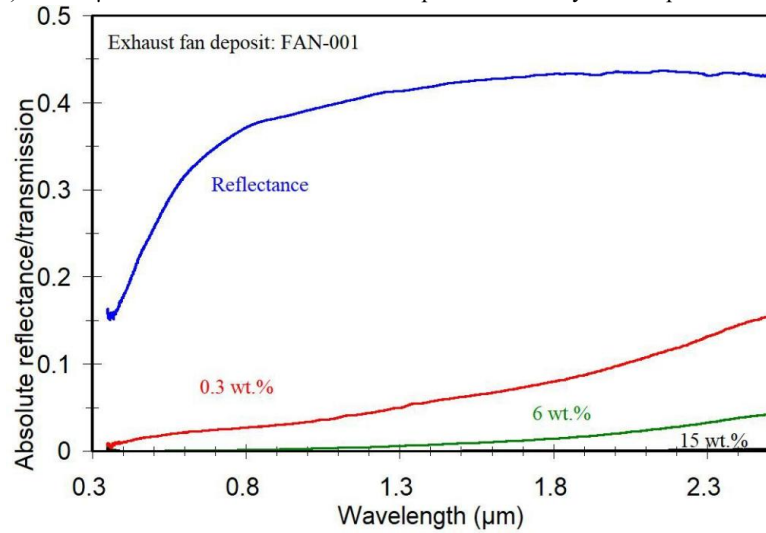
Figure 1. Cont.



(b) Same as Figure 1a for the 15 wt.% transmission spectrum.

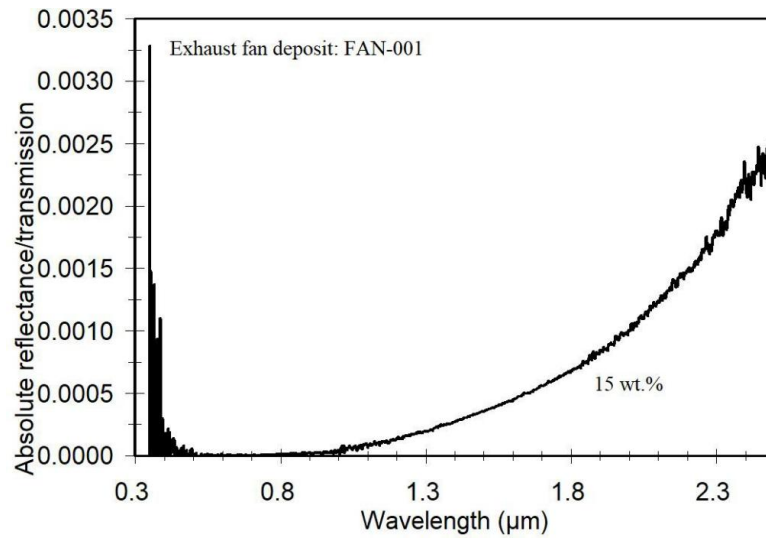


(c) 2.5–25 μm reflectance and transmission spectra of coal fly ash sample C-479994.

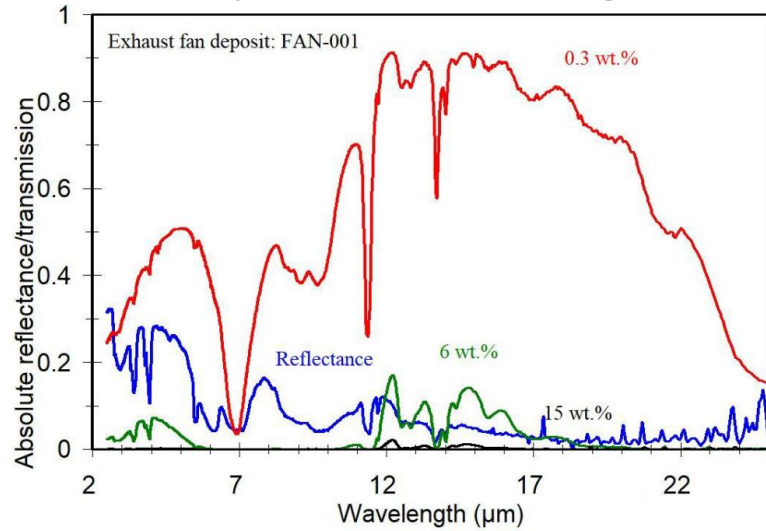


(d) 0.35–2.5 μm reflectance and transmission spectra of exhaust fan deposit FAN-001.

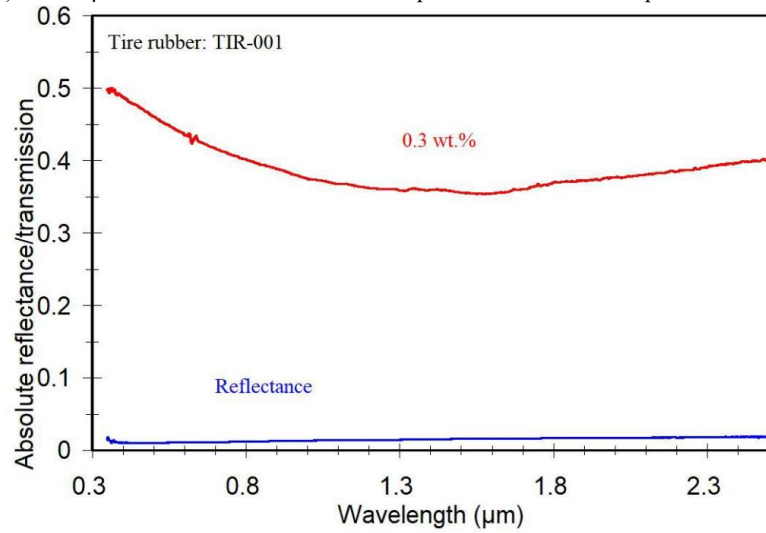
Figure 1. Cont.



(e) Same as Figure 1d for the 15 wt.% transmission spectrum.

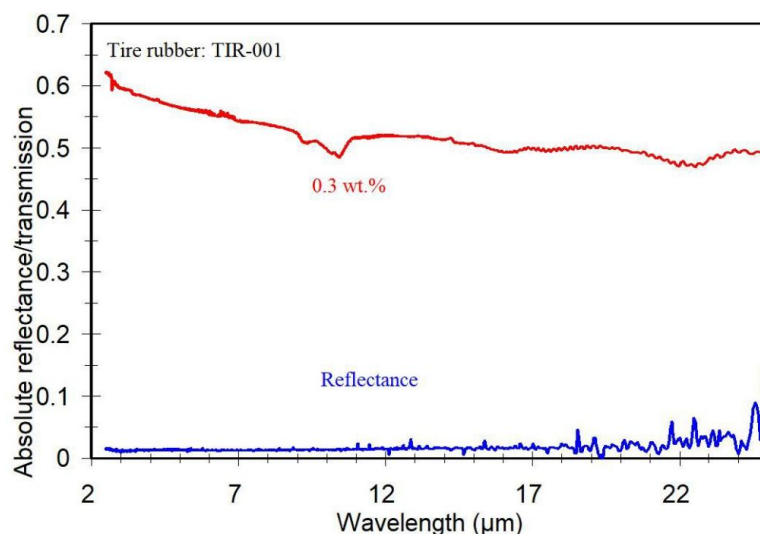


(f) 2.5–25 μm reflectance and transmission spectra of exhaust fan deposit FAN-001.



(g) 0.35–2.5 μm reflectance and transmission spectra of tire sample TIR-001.

Figure 1. Cont.



(h) 2.5–25 μm reflectance and transmission spectra of tire sample TIR-001.

Figure 1. Anthropogenic Dust Spectra.

In the 3–5 and 8–13 μm atmospheric windows (**Figure 1c**), this material shows a local reflectance maximum in the 3–5 μm region, and a major broad absorption band centered near 9.7 μm in reflectance, attributable to various Si-O and Al-O stretching fundamentals^[184–186]. In transmission, with increasing concentration, this longer wavelength band rapidly becomes saturated, resulting in transmission near 0%, and a broadening of the absorption feature, such that the region of high absorbance becomes wider. This has the effect of causing the lower wavelength transmission peak to move toward shorter wavelengths, as the absorption band widens and its wings extend to progressively shorter (and longer) wavelengths. As a result, by 6 wt.% concentration of the absorbing species, this spectral region is characterized by a broad region of high absorbance (**Figure 1c**). This is similar to the results seen in a previous study^[187], which examined the mid-infrared properties of three different fly ashes, all of which had similar XRF compositions to ours. This study found a single broad absorption feature, located around 9.71 μm (1030 cm^{-1}). The weak double feature on the edge of the transmission window at approximately 12.6 and 12.8 μm (795 and 779 cm^{-1}), is attributed to the quartz (and/or other silicates) in the original ash material.

3.1.2. Exhaust Fan Deposits (FAN-001) Spectra

The particulate matter off the parking garage intake-fan air filter lacked any discernible features in the 0.4–2.5 μm window (**Figure 1d–e**) but had several distinctive bands in

both the 3–5 and 8–13 μm transmission windows (**Figure 1f**). As expected for complex mixtures of organic-rich particles, absorbance is highest at lower wavelengths. Transmission for this sample increases by ~65% from 0.45 to 0.75 μm for the 0.3 wt.% sample spectrum and visible region reflectance is 2% at 0.56 μm . As concentration increases, transmission at all wavelengths decreases as expected, but the spectra remain red-sloped. Transmission for the 15 wt.% sample spectrum is <1%. No new absorption features appear with increasing concentration (**Figure 1d–e**).

The fan deposit appears to be a complex mixture of aluminosilicates, carbonates, chlorides, and organic compounds (**Table 2**). This likely accounts for its complex infrared spectrum (**Figure 1f**). Weak absorption features associated with carbonate minerals are seen between 3.33 and 3.51 μm (3000 and 2850 cm^{-1})^[188], with a likely additional contribution from C-H stretching of organics^[175]. Carbonate mineral absorption features are also seen between 3.70 and 4.00 μm (2700 and 2500 cm^{-1}). In the 8–13 μm window, numerous absorption bands contribute to the region between 8.33 and 10.93 μm (1200 and 915 cm^{-1}). Given the complex composition of the sample (**Table 2**) it is likely that these absorptions arise from contributions of silicate and carbonate minerals and overlapping organics. The sharp feature at 11.36 μm (880 cm^{-1}) is attributed to the out-of-plane-bend of CO_3^{2-} ^[188] and comes from the presence of carbonate in the sample, while the two features in the Si-O-Si symmetric

stretching region at 12.50 and 12.82 μm (800 and 780 cm^{-1}) are attributable to the presence of quartz and members of the albite-anorthite series^[184]. The strong absorption band near 6.9 μm is also attributable to carbonate^[188]. With increasing concentration, the various fundamental absorption bands in the 6–10 μm region become saturated, blending into a region of low (<1%) transmission.

3.1.3. Tire Rubber (TIR-001) Spectra

The tire rubber sample was difficult to process due to its elasticity, and did not produce readily identifiable absorption features in the 0.4–2.5 or 3–5 μm windows (**Figure 1g–h**). This is not unexpected for complex organic-rich materials. Transmission decreases slightly over the visible spectral region by ~15%, suggesting that use of a single optical constant would be reasonable for the visible spectral region.

Potentially useful features in the 8–13 μm window were found at approximately 9.26, 10.15 and 10.42 μm (1080, 985 and 960 cm^{-1}), with the first being attributed to symmetric C-O stretching and the latter two being attributed to C-H bends^[189] (**Figure 1h**).

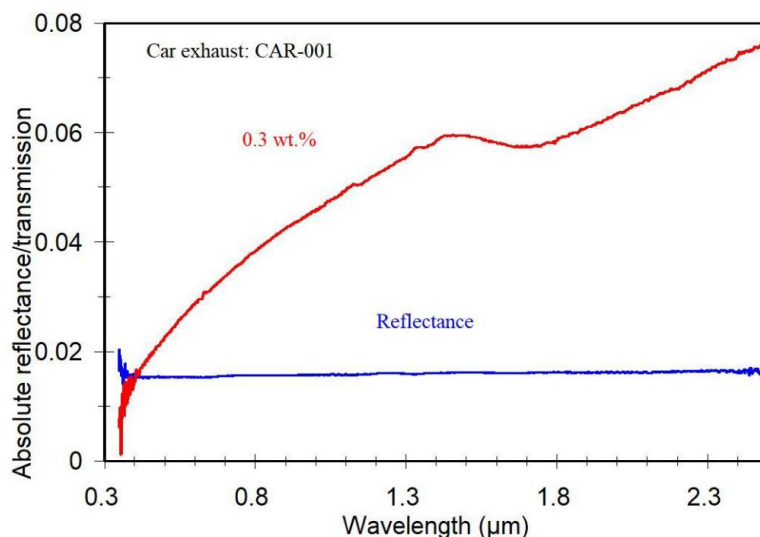
3.2. Carbonaceous Group Spectra

This group includes car exhaust deposits (sample CAR-001), and wood-burning chimney ash (CHA-001).

3.2.1. Car Exhaust (CAR-001) Spectra

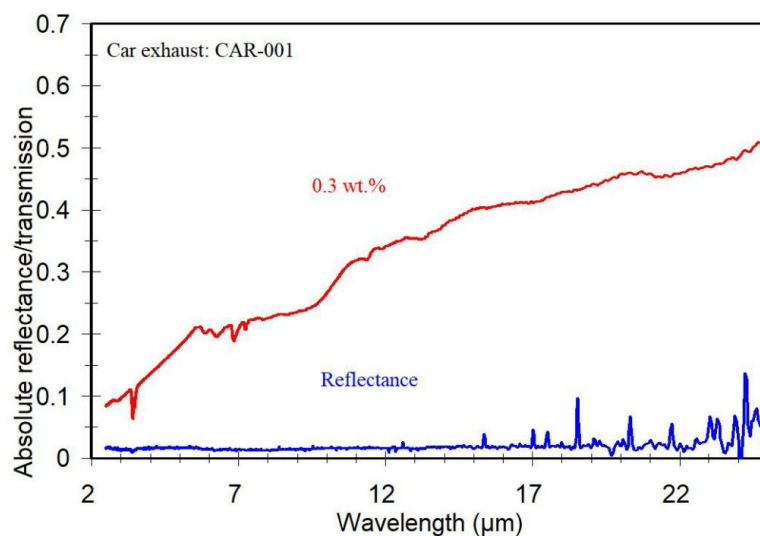
The car exhaust sample spectra lack sharp absorption features in the 0.4–2.5 μm region (**Figure 2a**). The bulk sample is dark (<2% reflectance) and spectrally flat and featureless. The 0.3 wt.% spectrum is strongly red-sloped as expected for complex mixtures of organics, similar to the exhaust fan particulates, but is even more red-sloped. Its 0.75/0.45 μm ratio is 1.86, with 0.56 μm reflectance of 2.6%. It exhibits a broad absorption feature centered near 1.7 μm , consistent with it containing a range of aliphatic organics^[190].

Absorption features appear in both the 3–5 and 8–13 μm windows (**Figure 2b**). In agreement with a previous study^[191] we see aliphatic C-H stretching in the 3.33–3.51 μm (3000–2850 cm^{-1}) region as well as the absence of the aromatic C-H absorptions at slightly higher frequencies, which would be expected in a diesel exhaust sample. The moderately deep feature in the 8.85–9.26 μm (1130–1080 cm^{-1}) is likely produced from overlapping absorptions of fundamental SO_4^{2-} bending, as well as multiple C-H and C-O absorptions^[175,192]. Two additional absorption features appear on the shoulder of the major band at approximately 11.30 and 11.90 μm (885 and 840 cm^{-1}), both of which are attributable to be produced by C-O stretches^[175]. Overall, the 0.3 wt.% spectrum shows increasing transmission toward longer wavelengths. It does not appear that any of the absorption bands in this region are saturated at this concentration.

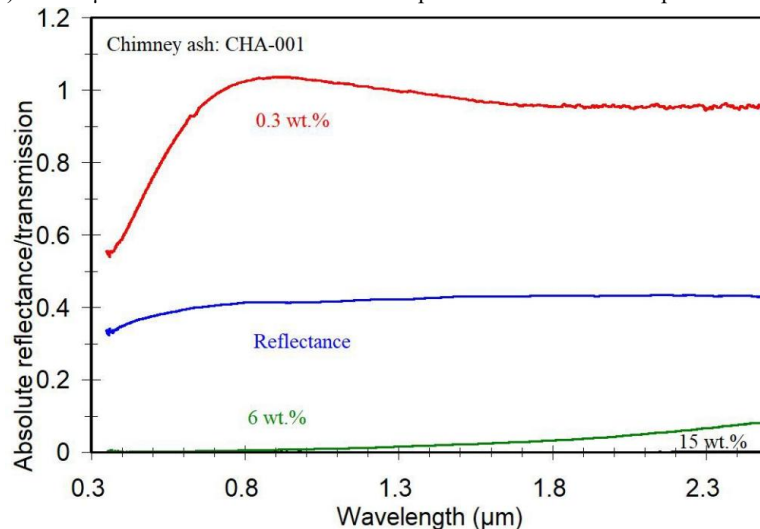


(a) 0.35–2.5 μm reflectance and transmission spectra of car exhaust sample CAR-001.

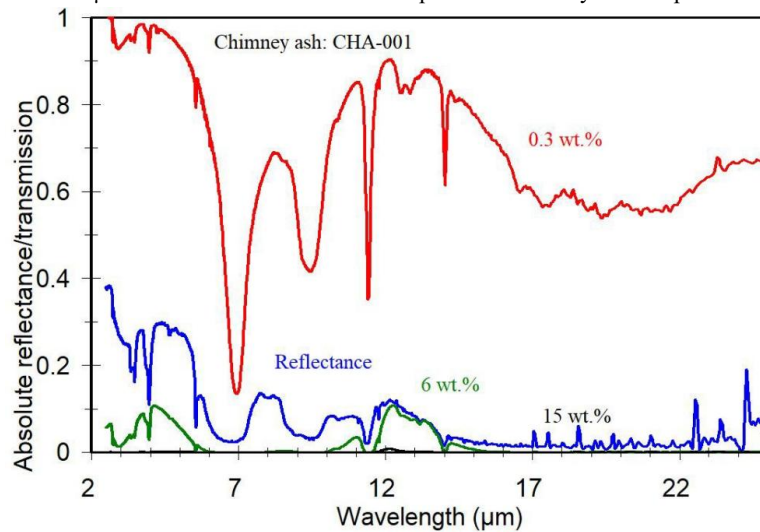
Figure 2. Cont.



(b) 2.5–25 μm reflectance and transmission spectra of car exhaust sample CAR-001.



(c) 0.35–2.5 μm reflectance and transmission spectra of chimney ash sample CHA-001.



(d) 2.5–25 μm reflectance and transmission spectra of chimney ash sample CHA-001.

Figure 2. Carbonaceous Group Spectra.

3.2.2. Chimney Ash (CHA-001) Spectra

The ash sample from the wood-burning fireplace is spectrally featureless over the 0.3–2.5 μm region (**Figure 2c**). The 0.3 wt.% spectrum shows low absorption with >80% transmission at 0.56 μm , transmission peaking near 0.85 μm , and red-sloped over the 0.35–0.85 μm interval. At 6 wt.% concentration, the spectrum becomes strongly red-sloped over the entire 0.35–2.5 μm interval. The 15 wt.% sample is nearly fully absorbing.

Weak absorption features in the 3–5 μm window are seen in the 3.33–3.57 μm (3000–2800 cm^{-1}) range, as well as at 3.85 and 3.98 μm (2595 and 2140 cm^{-1}) (**Figure 2d**). These features are mainly attributed to the remaining carbonate minerals in the sample (**Table 2**), with contributions from C-H stretching of aliphatic hydrocarbons in the 3.33–3.57 μm (3000–2800 cm^{-1}) range^[193]. The 8–13 μm window is dominated by the remaining inorganic materials in the sample, mainly silicates (9.35–9.50 μm region) and carbonates (7 and 11.5 μm regions). XRD results for the ash sample (**Table 2**) show that quartz and calcite likely make up the bulk of these minerals. A previous study^[193] found similar minerals in their samples (silicates and clays) and attributes their presence to the lower sensitivity of these materials to the heating that occurs during combustion compared to that of the organics. Weak features which we attribute to the Si-O-Si stretching of quartz are seen at 8.51, 8.70, 12.50 and 12.82 μm (1175, 1150, 800 and 780 cm^{-1}), with the first two being the asymmetric stretch and the latter being the symmetric stretches^[194]. The sharp absorption at 11.43 μm (875 cm^{-1}) is indicative of out-of-plane CO_3^{2-} bending in calcite^[188]. As with many of the other samples, increasing concentration leads to saturation and consequent broadening of the main absorption bands accompanied by continued presence of the weaker absorption bands, provided they fall outside the region of saturation. For this sample, increasing concentration does not result in the appearance of additional absorption bands (which may have been too weak to appear in the 0.3 wt.% spectrum), but some absorption bands broaden as increasing sample concentration leads to band saturation. Absorption band saturation also leads to some “smearing out” of adjacent absorption bands as they reach saturation

(e.g., 6–10 μm region of the 15 wt.% sample (**Figure 2d**).

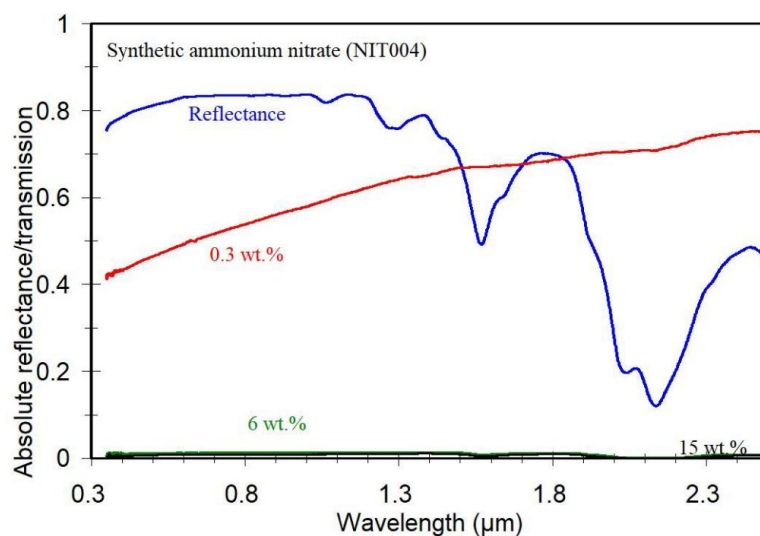
3.3. Secondary Group Spectra

This group includes synthetic ammonium nitrate (NIT004) and two samples of ammonium sulfate, one naturally-occurring (SPT101; mineral name mascagnite) and a synthetic one (SPT105).

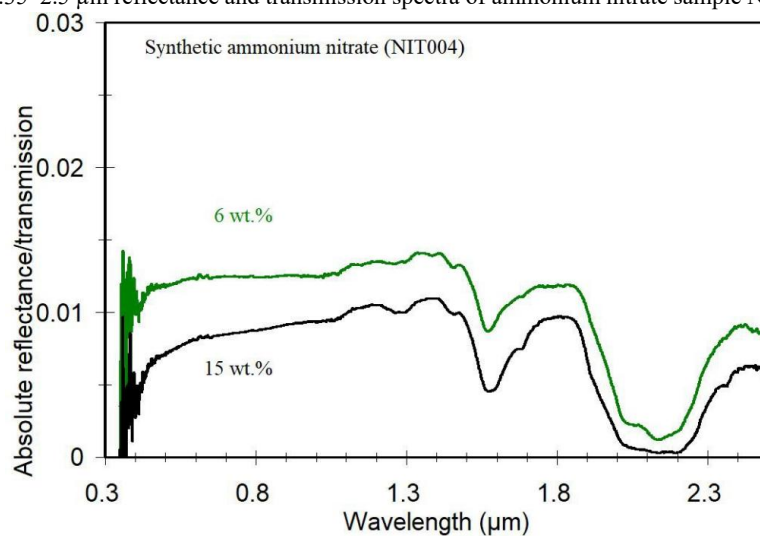
3.3.1. Synthetic Ammonium Nitrate (NIT004) Spectra

The synthetic ammonium nitrate used in this study shows strong absorption features in the 0.4–2.5 μm region, particularly near 1.55 and 2.0–2.15 μm (**Figure 3a,b**). These bands, and others are attributable to overtones and combinations of NH_4 fundamentals^[179]. The spectrum is slightly red-sloped in the visible region (0.75/0.45 μm reflectance ratio of 1.17) for the 0.3% sample spectrum, and becomes bluer for the higher-concentration sample spectra. The 6 and 15 wt.% transmission spectra clearly exhibit the ammonium-associated absorption features, even though overall transmission is low (<1.5%) (**Figure 3b**). The longer wavelength absorption feature is broadened due to saturation. An overtone/combination feature near 1.29 μm is most evident in the 15 wt.% sample spectrum (**Figure 3b**).

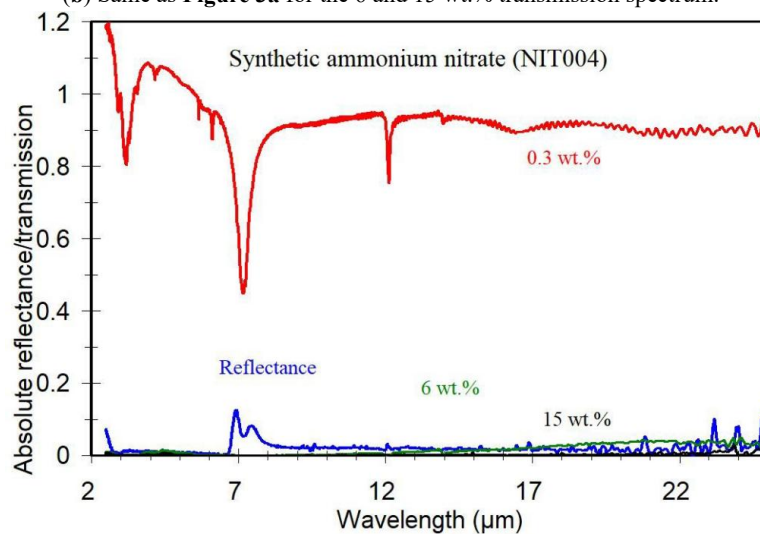
The 3–5 μm window is dominated by various vibrational modes of the NH_4^+ molecule (**Figure 3c**). Absorption features in this region occur at approximately 3.01–3.19 μm (3135–3322 cm^{-1}), 3.2–3.30 μm (3030–3125 cm^{-1}) and 3.45–3.57 μm (2805–2900 cm^{-1})^[195]. These features are only visible in the 0.3% concentration sample, as the higher concentration spectra have near total absorbance. There is an additional weak feature partially masked by the presence of some CO_2 at 4.17 μm (2400 cm^{-1}) that is potentially the third overtone of the out-of-plane bend of NO_3^- . The 8–13 μm window consists of two absorption features, a symmetric stretch and an out-of-plane deformation of the NO_3^- molecule at 9.62 and 12.12 μm (1040 and 825 cm^{-1}) respectively^[195]. In contrast to most of the other samples, the 9.62 μm stretching vibration only appears weakly in the 6% concentration pellet; however, total absorption across the entire spectrum is extremely high.



(a) 0.35–2.5 μm reflectance and transmission spectra of ammonium nitrate sample NIT004.

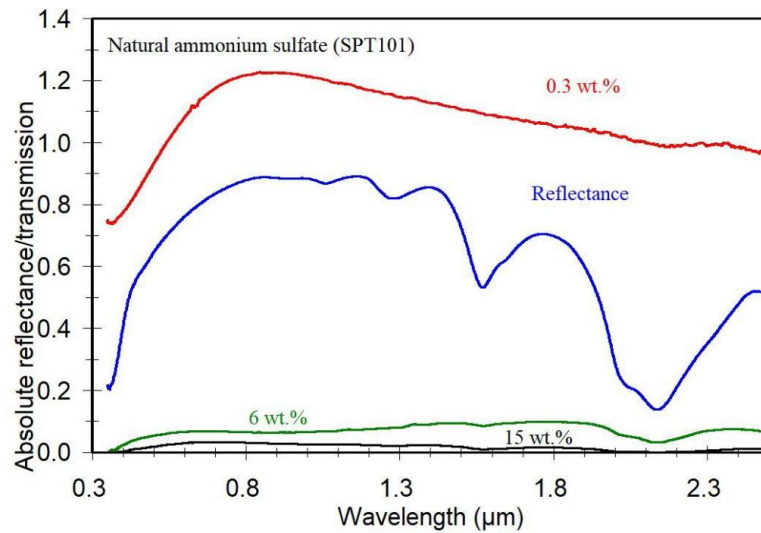


(b) Same as **Figure 3a** for the 6 and 15 wt.% transmission spectrum.

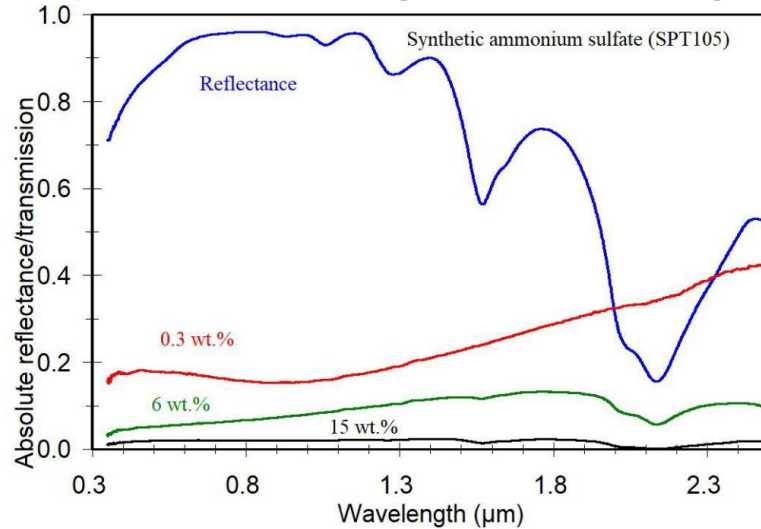


(c) 2.5–25 μm reflectance and transmission spectra of ammonium nitrate sample NIT004.

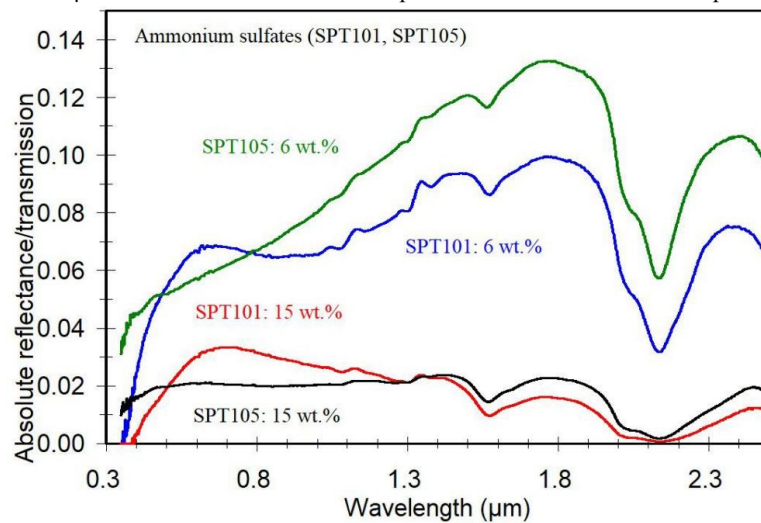
Figure 3. *Cont.*



(d) 0.35–2.5 μm reflectance and transmission spectra of ammonium sulfate sample SPT101.

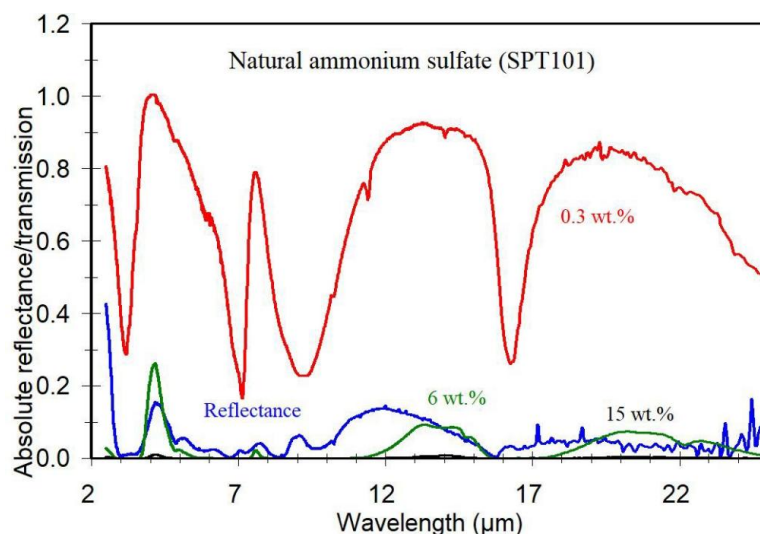
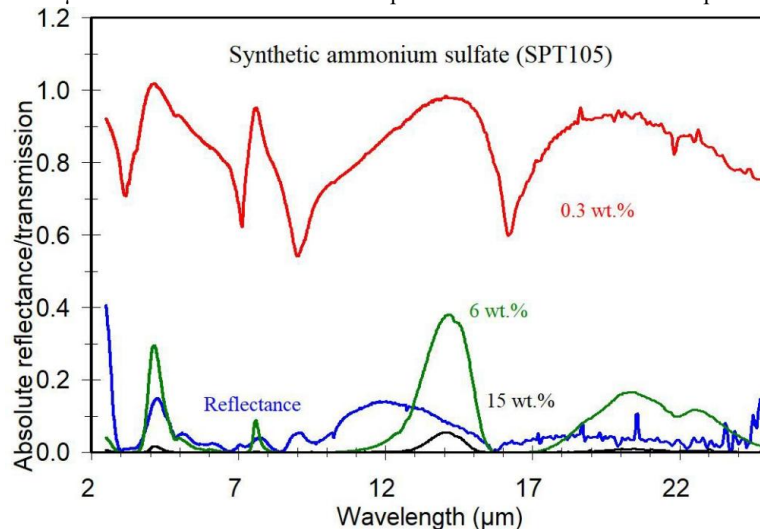


(e) 0.35–2.5 μm reflectance and transmission spectra of ammonium sulfate sample SPT105.



(f) 0.35–2.5 μm transmission spectra of two ammonium sulfate samples (SPT101 and SPT105) for 6 and 15 wt.% samples.

Figure 3. Cont.

(g) 2.5–25 μm reflectance and transmission spectra of ammonium sulfate sample SPT101.(h) 2.5–25 μm reflectance and transmission spectra of ammonium sulfate sample SPT105.**Figure 3.** Secondary Group Spectra.

3.3.2. Ammonium Sulfates (SPT101 and SPT105) Spectra

We investigated the spectral transmission properties of ammonium sulfate using both a natural sample (SPT101) which contains some tschermigite (an aluminum-ammonium sulfate) as well as reagent-grade ammonium sulfate (SPT105). The two bulk powder reflectance spectra are similar (**Figure 3d–e**). Like the ammonium nitrate (NIT004), both spectra are dominated by strong N-H absorption bands in the 1.55 and 2.15 μm regions^[179]. At 0.3 wt.% concentrations, the 0.35–2.5 μm spectra have different shapes and absolute reflectance. The cause of these differences is not fully understood, but may relate to differences in composition. Both show a weak absorption feature in the 2.15 μm

region, while the SPT105 spectrum also exhibits a broad absorption feature centered near 0.9 μm , likely due to small amounts of iron. The 6 wt.% samples provide the best opportunity for identifying spectral signatures in this region for both samples (**Figure 3f**), and their spectra show fewer differences than the 0.3 wt.% samples. Combinations of the stretching and bending modes of the NH_4^+ molecule dominate the region for both samples, with approximate band locations at 1.56, 2.03 and 2.14 μm ^[179]. While the presence of these features can be seen in the 15% concentration samples, the 2 μm region absorption feature is partially saturated, with a broadening of this feature and loss of band position details. Higher order N-H bands near 1.1 and 1.3 μm are more apparent in the 15 wt.% sample spectra.

The 3–5 and 8–13 μm regions show strong absorption bands at 0.3 wt.% concentration (**Figure 3g–h**), in contrast to the lower wavelength spectra. In the 3–5 μm window absorption features found at approximately 3.18 μm (3140 cm^{-1}), 3.30 μm (3030 cm^{-1}), and 3.55 μm (2820 cm^{-1}) are again attributed to the various overtone and combination modes of NH_4^+ molecule^[179,195]. These features, however, only appear well-resolved in the 0.3% concentration spectra, as near absolute absorption occurs at higher concentrations. The 8–13 μm window is dominated by overlapping modes of the SO_4^{2-} molecule. In this region the synthetic and natural sample's absorption feature shapes appear to deviate, as the natural (mascagnite) sample has a much broader absorption between 8.33 and 9.80 μm (1020 and 1200 cm^{-1}), while the synthetic sample produced a flatter appearance, with a more discrete band minimum at 9.01 μm (1110 cm^{-1}). These features are assigned to the asymmetric bending vibration of SO_4^{2-} ^[102]. A weak shoulder feature appears at 10.26 μm (975 cm^{-1}) which may be the SO_4^{2-} symmetric stretching vibration^[180]. The mascagnite sample (SPT101) has an additional sharp absorption feature at 11.42 μm (875 cm^{-1}) that is absent in the synthetic sample, likely due to a minor amount of carbonate in this sample. An S-O overtone absorption feature appears on the edge of the transmission window at 4.83 μm (2070 cm^{-1}). As with many of the other samples, the 0.3 wt.% concentration spectra are adequate to show all the diagnostic absorption features in the 3–5 and 8–13 μm windows, fundamental absorption bands are not saturated, and increasing concentration rapidly leads to saturation and widening of them. The higher concentration samples allow some minor features, such as the 4.83 μm absorption band, which is weak in the 0.3 wt.% spectrum, to become more apparent. NH_4 -associated absorption band positions are not very different between ammonium sulfate and ammonium nitrate^[179].

3.4. Mineral Group Spectra

Along with rocks, this is the largest group of samples examined and includes iron carbonate (siderite; CRB145), illite (phyllosilicate; ILL001), kaolinite (phyllosilicate; KAO101), potassium feldspar (KFE001), quartz (QUA101), and calcium sulfate (gypsum; SPT178).

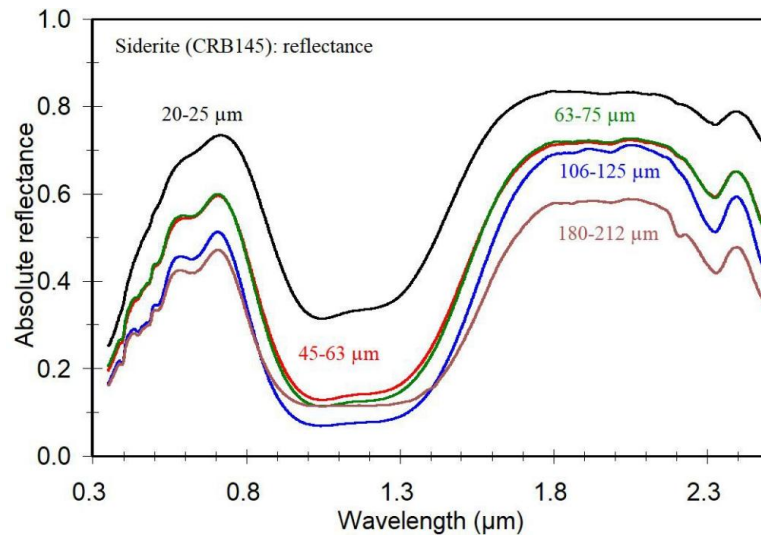
3.4.1. Carbonate (Siderite; CRB145) Spectra

We selected siderite, an iron-rich carbonate to investigate in more detail how grain size affects transmission spectra, as well as how reflectance and transmission spectra compare. Carbonates are a significant component of atmospheric dust from many sources^[99,196,197].

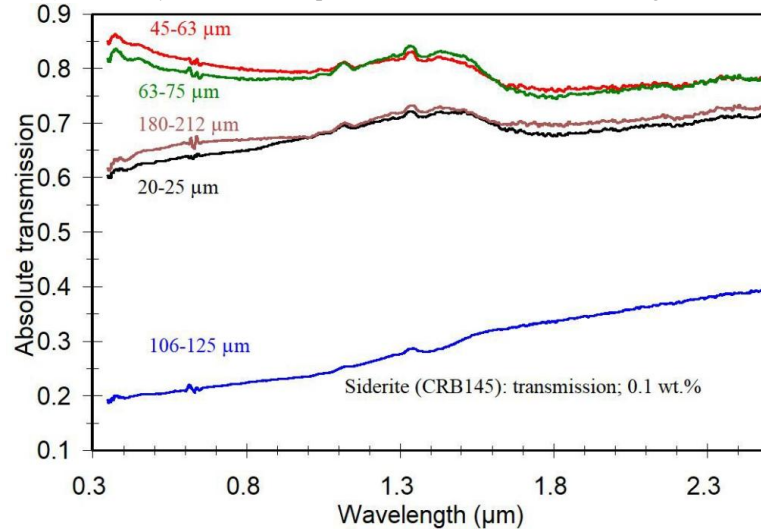
The 0.3–2.5 μm reflectance spectra (**Figure 4a**) all show Fe^{2+} -associated absorption features in the 0.4–0.6 μm region (series of narrow absorption bands) and 1–1.3 μm regions (doublet—partially overlapping absorption bands centered near 1.0 and 1.25 μm), and C-O associated overtone and combination bands near 2.17 and 2.32 μm ^[198]. With increasing grain size, the short wavelength absorption bands become stronger and more apparent, as do the longer wavelength carbonate bands. The two 1- μm region Fe^{2+} absorption bands tend to blend together, due to increasing band saturation.

In transmission, the 0.1 wt.% samples (**Figure 4b**) are weakly featured, with generally high transmittance except for the second-largest grain size sample. The 1- μm region Fe^{2+} absorption band is only weakly evident in the two smallest grain size spectra. Narrow features in the 1.1–1.4 μm region are spectral artifacts. The 1 wt.% spectra (**Figure 4c**) show variable evidence for the 1- μm region absorption band in the smaller size fractions, as well as the aforementioned spectral artifacts. There is no systematic variation in overall transmission, likely because of incomplete mixing of the sample with KBr. We used minimal mechanical mixing in order to preserve the grain size of the minerals in each pellet.

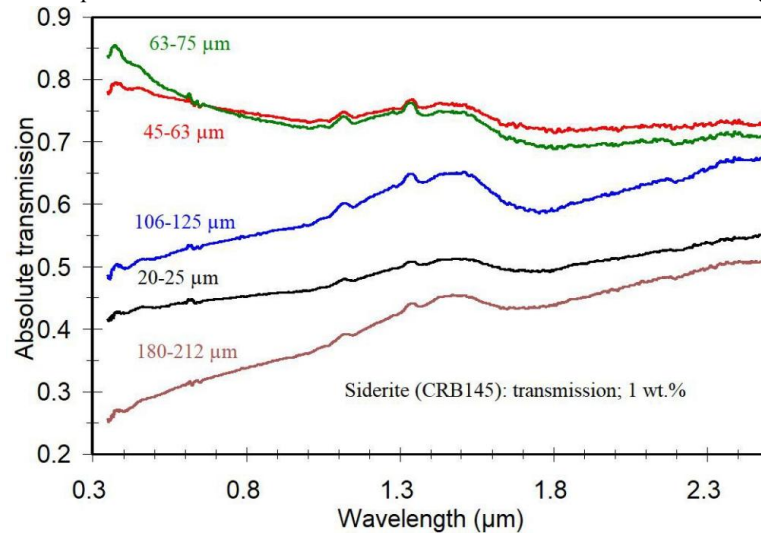
The infrared reflectance spectra (**Figure 4d**) have a number of absorption features associated with C-O bending and stretching fundamental vibrations, as well as various overtones and combinations^[188]. Overall reflectance generally decreases with increasing grain size as expected. The corresponding IR spectra for 0.1 wt.% siderite (**Figure 4e**) show variable spectral behavior. An absorption band is seen in the finest grain size spectra near 7 μm (C-O band)^[188], but the spectra are otherwise featureless. Most of the spectra are quite flat. The 1 wt.% spectra show more spectral features (**Figure 4f**), again for the finest fractions. In these spectra, the 7 μm feature is evident, as well as other C-O absorption bands^[188], particularly near 11.5 and 13.5 μm .



(a) 0.35–2.5 μm reflectance spectra of CRB145 as a function of grain size.

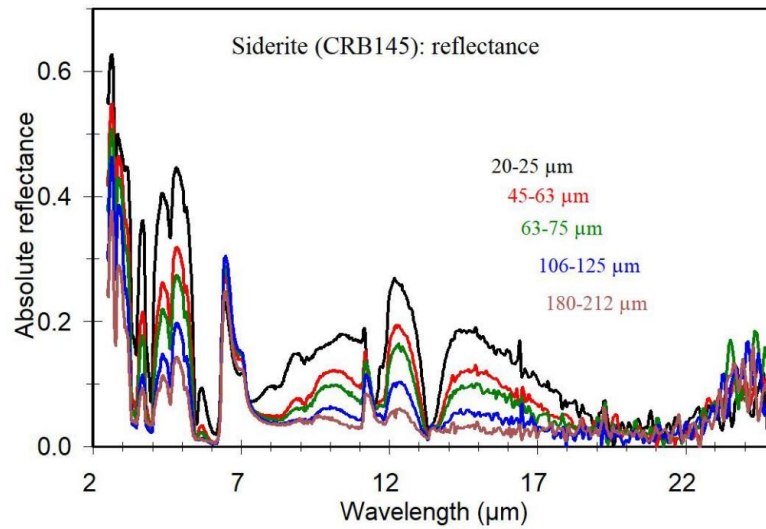


(b) 0.35–2.5 μm transmission spectra of carbonate CRB145 for 0.1 wt.% concentrations and different grain sizes (as indicated).

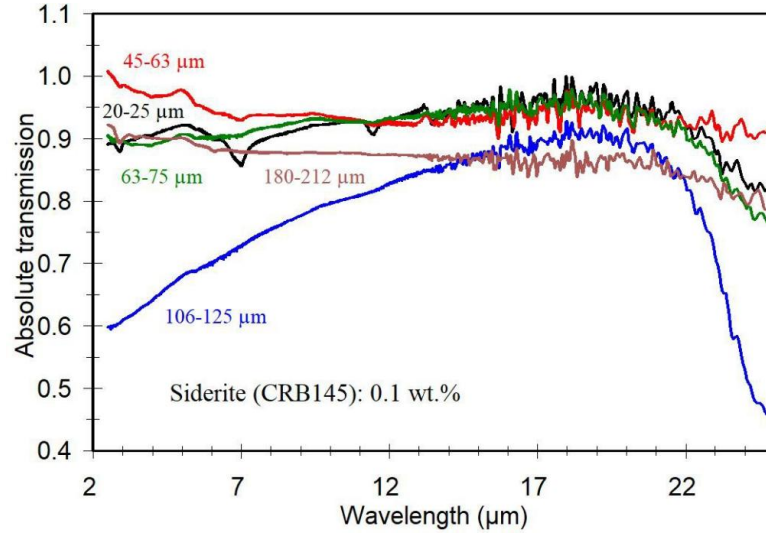


(c) 0.35–2.5 μm transmission spectra of carbonate CRB145 for 1 wt.% concentrations and different grain sizes (as indicated).

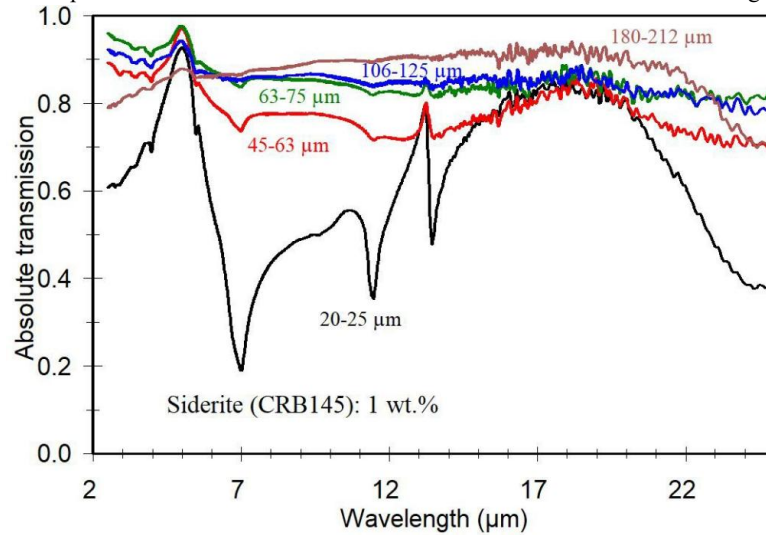
Figure 4. Cont.



(d) 2.5–25 μm reflectance spectra of carbonate CRB145 for different grain sizes (as indicated).

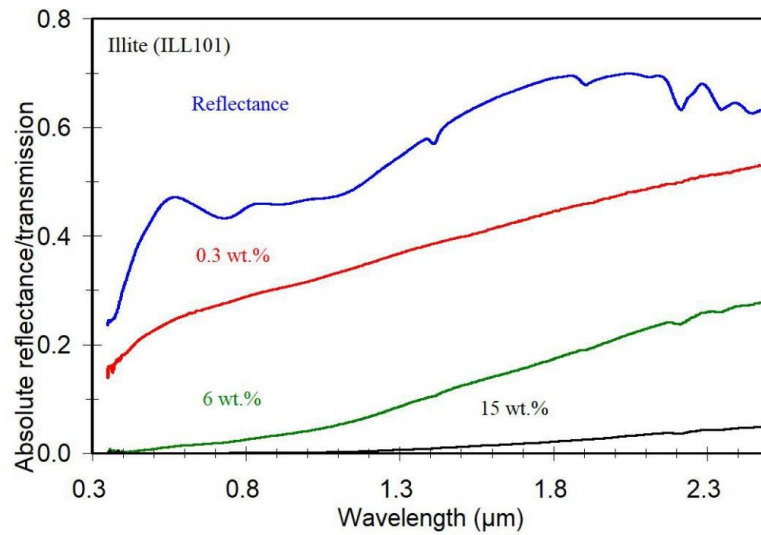


(e) 2.5–25 μm transmission spectra of carbonate CRB145 for 0.1 wt.% concentrations and different grain sizes (as indicated).

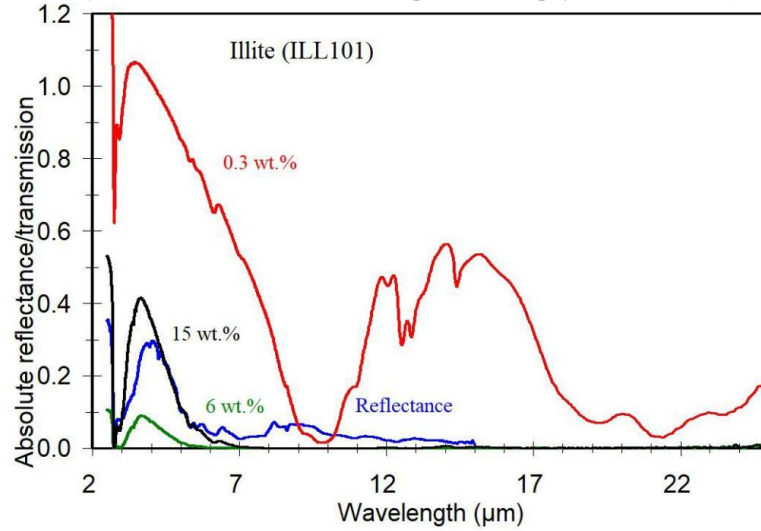


(f) 2.5–25 μm transmission spectra of carbonate CRB145 for 1 wt.% concentrations and different grain sizes (as indicated).

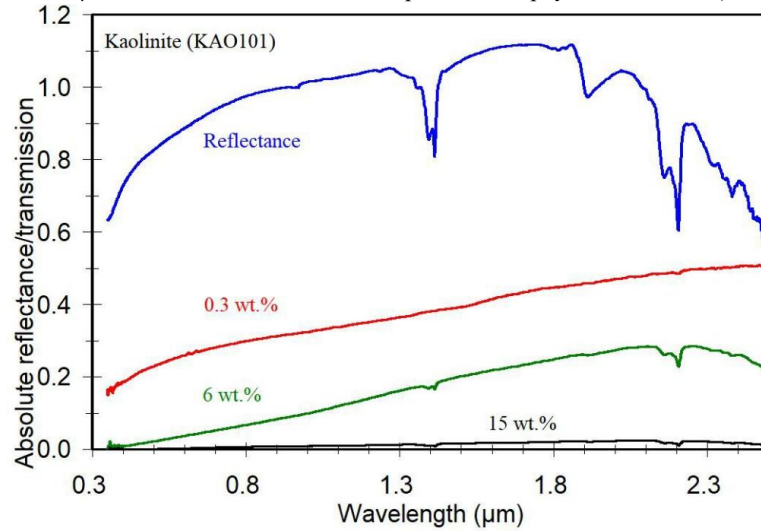
Figure 4. Cont.



(g) 0.35–2.5 μm reflectance and transmission spectra of the phyllosilicate illite (ILL101).

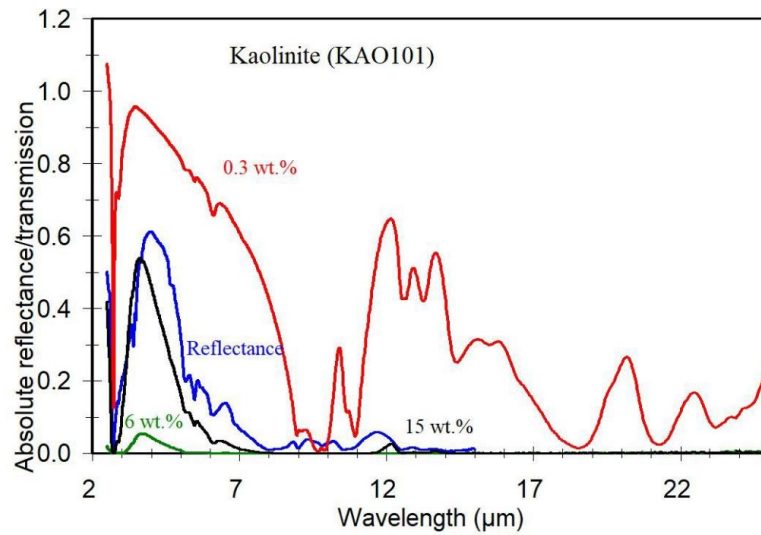


(h) 2.5–25 μm reflectance and transmission spectra of the phyllosilicate illite (ILL101).

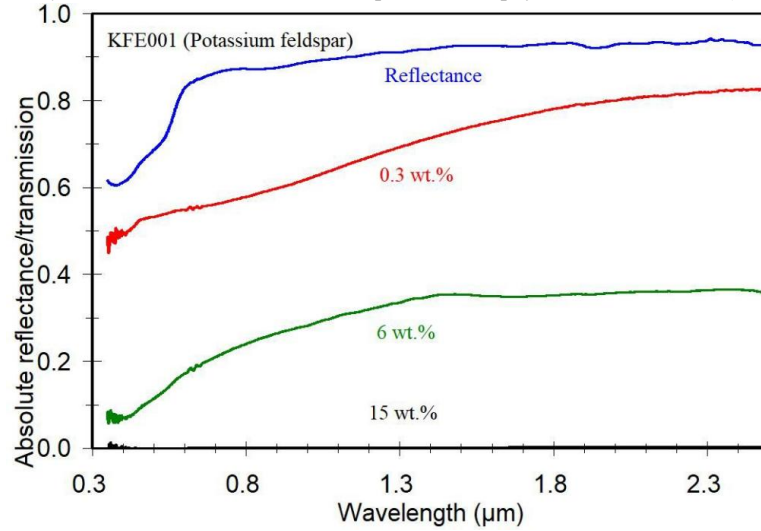


(i) 0.35–2.5 μm reflectance and transmission spectra of the phyllosilicate kaolinite (KAOL101).

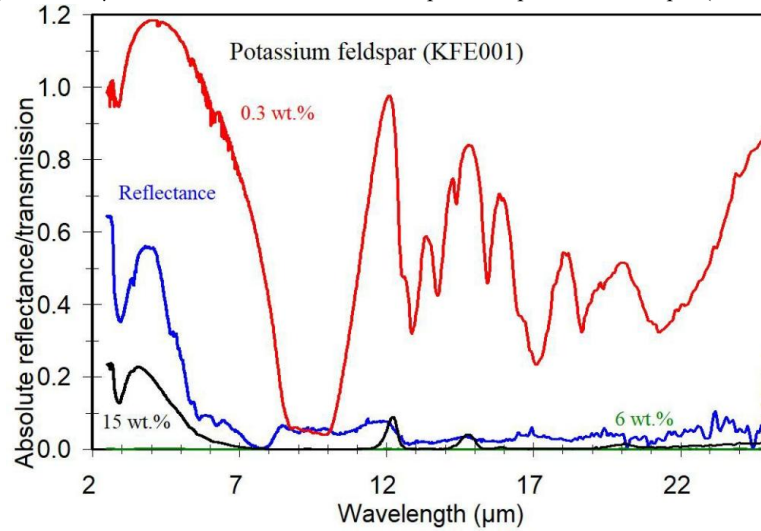
Figure 4. Cont.



(j) 2.5–25 μm reflectance and transmission spectra of the phyllosilicate kaolinite (KAO101).

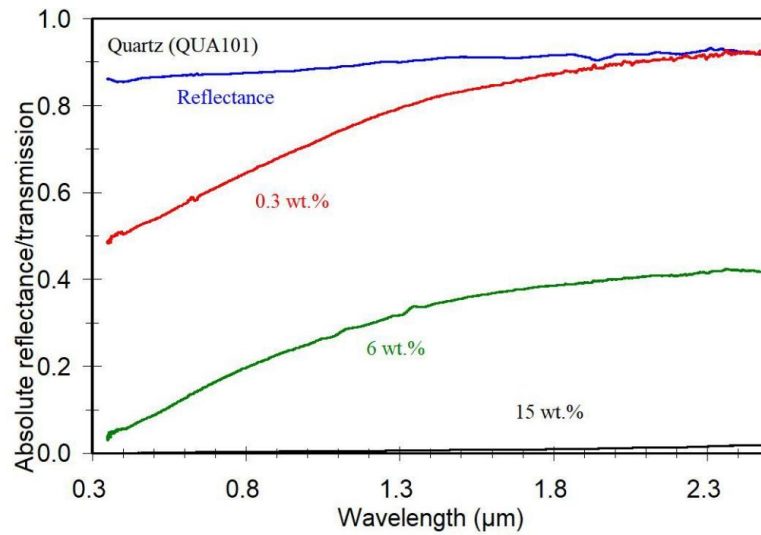


(k) 0.35–2.5 μm reflectance and transmission spectra of potassium feldspar (KFE001).

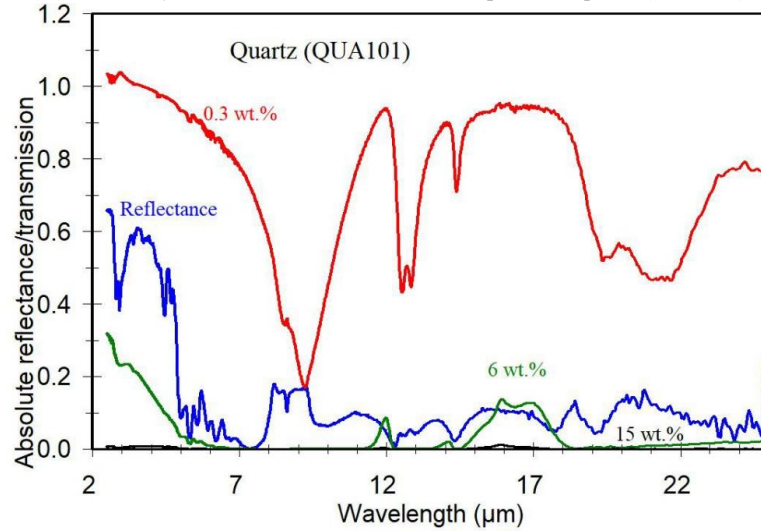


(l) 2.5–25 μm reflectance and transmission spectra of potassium feldspar (KFE001).

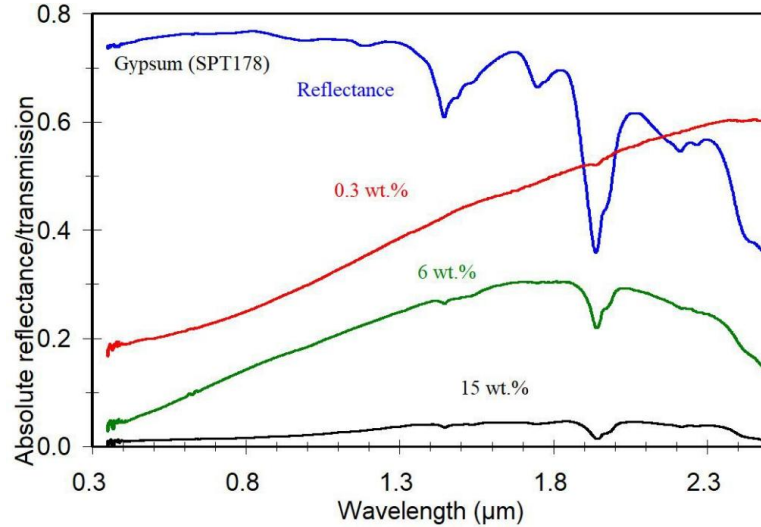
Figure 4. Cont.



(m) 0.35–2.5 μm reflectance and transmission spectra of quartz (QUA101).

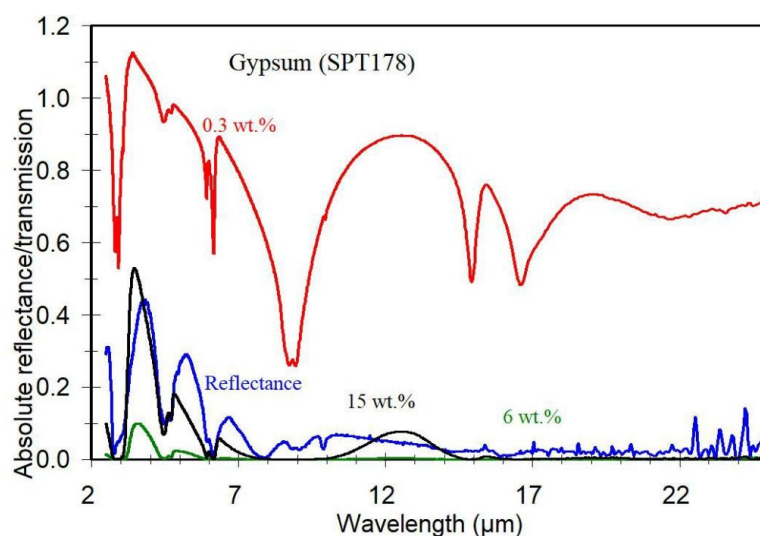


(n) 2.5–25 μm reflectance and transmission spectra of quartz (QUA101).



(o) 0.35–2.5 μm reflectance and transmission spectra of gypsum (SPT178).

Figure 4. Cont.



(p) 2.5–25 μm reflectance and transmission spectra of gypsum (SPT178).

Figure 4. Mineral Group Spectra.

3.4.2. Illite (Phyllosilicate; ILL101) Spectra

Illite (a phyllosilicate) is an important component of atmospheric dust in many areas^[199–201]. It is a complex phyllosilicate with the formula $\text{K}_{0.6–0.85}\text{Al}_2(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2$ or even $\text{K}_{0.6–0.85}(\text{Al},\text{Mg})_2(\text{Si},\text{Al})_4\text{O}_{10}(\text{OH})_2$, allowing extensive elemental substitutions. Illites are also often poorly crystalline. The 0.35–2.5 μm reflectance spectrum (**Figure 4g**) shows a number of weak absorption features: an Fe^{3+} – Fe^{2+} charge transfer band near 0.75 μm , two Fe^{2+} crystal field transition bands near 0.9 and 1.1 μm , an OH absorption band near 1.4 μm , an H_2O absorption band near 1.9 μm , and metal-OH bands from 2.2–2.45 μm ^[176].

While mostly absent in the 0.3% concentration sample spectrum (**Figure 4g**), the metal-OH bands in the 2.2–2.45 μm region are weakly present in the 6 and 15 wt.% sample spectra. All the spectra are red-sloped, and the transmission spectra are increasingly dark with increasing concentration.

Absorption features are largely absent in the 3–5 μm region with the exception of OH and H_2O fundamental stretching bands in the 2.7–3.3 μm region (**Figure 4h**). The broad feature in the 8–13 μm window is composed of overlapping absorption features associated with Al-O and Si-O stretches of illite and possibly quartz, which is also present in this sample (**Table 3**). Absorption bands seen at 8.58, 9.17, and 9.85 μm (1165, 1090 and 1015 cm^{-1}) are attributable to asymmetric Si-O-(Si,Al) stretching. OH deformation features are seen at 10.93 and 12.05 μm (915 and 830 cm^{-1}), being Al-Al-OH and Al-Mg-OH respectively^[183,202]. The symmetric Si-O-Si

stretching typical of quartz is seen at 12.50 and 12.82 μm (800 and 780 cm^{-1}).

3.4.3. Kaolinite (Phyllosilicate; KAO101) Spectra

As with illite, kaolinite is another phyllosilicate that is abundant in atmospheric dust in many regions^[200,201,203]. The reflectance spectrum (0.35–2.5 μm) of kaolinite (**Figure 4i**) is characterized by only a few potentially diagnostic absorption bands, in the 1.4 and 2.1–2.3 μm region, attributable to OH overtones and SiOH combinations, respectively^[179]. These bands become more apparent in the higher concentration transmission spectra, particularly near 1.4 and 2.2 μm . The transmission spectra become increasingly darker and red-sloped, up to ~ 2 μm , with increasing concentration.

Diagnostic absorption features are absent in the 3–5 μm region, as expected (**Figure 4j**). The absorption feature in the 2.7–3.3 μm region is attributable to OH/ H_2O and is generally non-diagnostic of a particular species^[179,204]. The 8–13 μm region has a complex array of overlapping bands, which saturate and merge at higher dust concentrations. The presence of both Al and Si in the kaolinite structure (ideal formula: $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$) causes some distortions in the asymmetric Si-O stretching region (950–1200 cm^{-1}) that are also seen in some of the other silicate samples in this study. Two additional bands with transmission minima around 10.6 and 10.9 μm (940 and 915 cm^{-1}) are associated with OH deformation of inner-surface hydroxyl groups and OH deformation of inner hydroxyl groups respectively^[205]. As compared to

illite (see above), the kaolinite spectra show better-resolved individual absorption bands.

3.4.4. Potassium Feldspar (KFE001) Spectra

Potassium feldspar is a significant component of atmospheric dust^[200] and may serve as an important ice nucleating species^[206–209]. Its reflectance spectrum is largely featureless due to its lack of strongly absorbing atoms and molecules. (**Figure 4k**). The transmission spectra are modestly red-sloped and largely featureless, with transmission decreasing with increasing concentration. The reflectance/transmission dropoff towards the ultraviolet, and weak band near 0.8 μm are attributable to trace amounts of iron in the sample.

In the 8–13 μm window there are overlapping asymmetric Si-O-(Si,Al) fundamental stretches which result in a region of strong, nearly saturated absorption over the ~8.5–10 μm interval, and a lack of distinguishable band minima is seen (**Figure 4l**). Additionally, the typical Si-O-(Si,Al) symmetric stretching vibrations are seen, although the typical shape is lost as the higher frequency band, usually seen at 12.50 μm (800 cm^{-1}), is more of a shoulder feature in this sample spectrum. This leads to a slight shift in position to 12.57 μm (795 cm^{-1}), while the second band retains its position at 12.82 μm (780 cm^{-1}).

3.4.5. Quartz (QUA101) Spectra

Quartz is present in many atmospheric dust samples^[210–212], as it is a common and widespread mineral on Earth. In the 0.4–2.5 μm window, the quartz spectra are nearly featureless, regardless of concentration (**Figure 4m**). The transmission spectra are modestly red-sloped. This is not unexpected and the only discernible expected absorption feature is an Si-OH doublet in the 2.20–2.27 μm region^[177]; it is only weakly seen in the reflectance spectrum.

In the 3–5 μm window the 0.3% sample spectrum did not exhibit usable absorption features, however, at increasing concentrations a number of features become apparent though total transmission is low (**Figure 4n**). Features in this region are assigned to the various overtones and combinations of Si-O, with bands apparent at 3.97, 4.47 and 4.67 μm (2520, 2235 and 2140 cm^{-1})^[185]. In the 8–13 μm window, absorption features in the 6 and 15% samples are lost due to near total light absorption, while the 0.3% concentration shows a number of absorption bands. Three absorption features together, derived from asymmetric Si-O stretching, domi-

nate the region, with bands located at 8.55, 8.73 and 9.25 μm (1170, 1145 and 1080 cm^{-1}). Additionally, symmetric Si-O-Si stretching produces a double feature with bands at 12.50 and 12.82 μm (800 and 780 cm^{-1}).

3.4.6. Gypsum (SPT178) Spectra

Gypsum is another common mineral component of mineral dust, and may be the dominant species in some areas^[213–216]. Gypsum may form through various processes in addition to being directly sourced from the Earth's surface^[217,218].

The reflectance spectrum of gypsum (0.35–2.5 μm) shows multiple absorption bands attributable to its structural water and S-O absorption features^[192]. The shapes and wavelength positions of the water-associated absorption bands are diagnostic of gypsum. The transmission spectra collected for gypsum in the 0.35–2.5 μm region showed only weak absorption features at 0.3 wt.% concentration and an overall red-sloped spectrum (**Figure 4o**) increasing in strength at 6% and 15% concentration. Unfortunately, most of these features fall within or on the edge of atmospheric “dead zones” as they are attributable to H₂O overtones and combinations (1.47, 1.52, 1.94 and 1.96 μm)^[192]. Additionally, weak features at 2.21 and 2.27 μm are seen, which are attributable to either combinations and/or overtones of OH/H₂O or the third overtone of S-O asymmetric bending vibrations, but they are extremely weak, even at 15 wt.% concentration. On the edge of the transmission window two features attributed to overtones of the S-O asymmetric bending vibration can be seen at 2.43 and 2.48 μm , but they significantly overlap with other absorption bands. The details of these longer wavelength bands are generally lost in the transmission spectra, reducing their utility for gypsum identification.

The 3–5 μm window is dominated by two distinct regions of absorption features (**Figure 4p**). First, the broad shoulder starting around 3.22 μm (3100 cm^{-1}) leading to the major H₂O stretching fundamentals at shorter wavelengths, can be seen at the edge of the atmospheric transmission window. This feature becomes broader and more saturated at increased concentrations. The second set of features is attributable to SO₄ overtones and combinations, with major bands appearing around 4.45 and 4.72 μm (2245 and 2120 cm^{-1}). The 8–13 μm window is dominated by SO₄ asymmetric bends, with band minimums around 8.73 and 8.93 μm (1145 and 1120 cm^{-1}). A very weak feature is also present

on the shoulder of this main feature which is attributable to the symmetric stretching of the SO_4 molecule^[180,219]. As expected, the 15% sample spectrum shows broad regions of saturation. None of the bands appears saturated in the 0.3% sample spectrum. Overall, it appears that gypsum is readily detectable over a range of concentrations because it possesses a large number of absorption features, most of which are detectable at low concentrations and do not become saturated until concentrations exceed many percent.

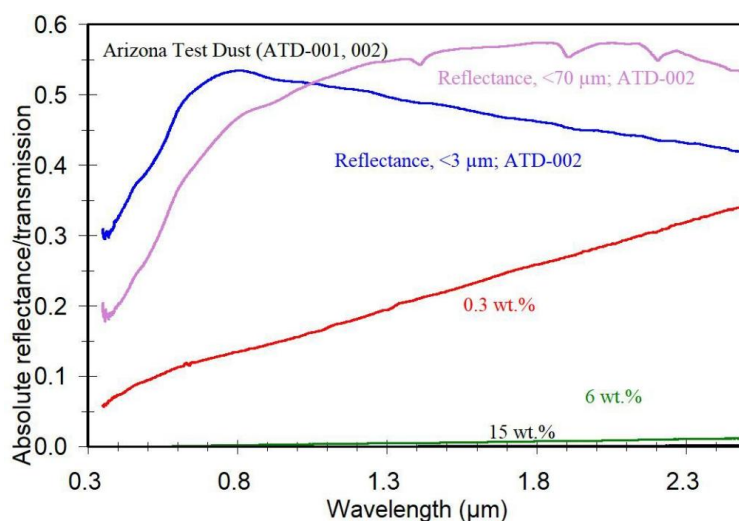
3.5. Rock/Mineral Mixture Group Spectra

Along with minerals, this is the largest group of samples examined and includes Arizona Test Dust (ATD-001, 002), loess (LOE-001), volcanic ash (Mt. St. Helens; MSH-001),

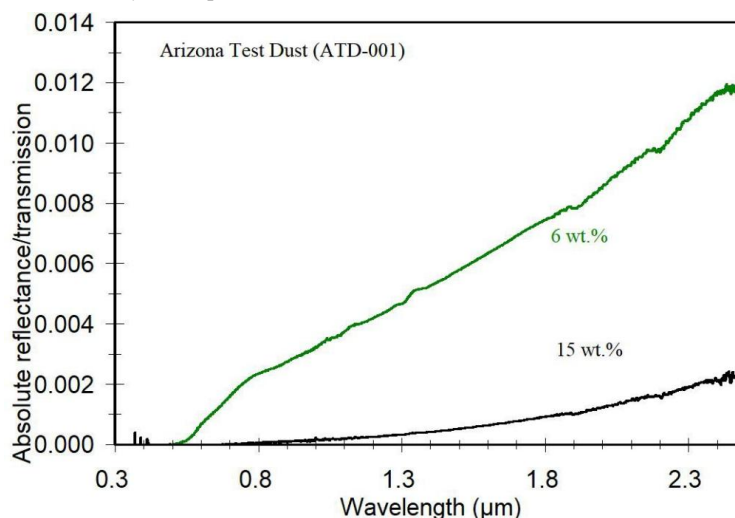
and rhyolite (RHY001).

3.5.1. Arizona Test Dust (ATD-001, ATD-002)

Arizona Test Dust (ATD) is a widely-used standard for various industrial applications^[220,221]. It is composed predominantly of quartz (49%), K-feldspar (15%), albite (15%), illite (11%), calcite (6%) and kaolinite (4%)^[222]. As such, its spectral characteristics are similar to the other major silicate-bearing samples used in this study, particularly in the 0.4–2.5 and 8–13 μm windows (**Figures 5a–c**). The two bulk powder spectra (<3 μm : ATD-001; <70 μm : ATD-002) differ in terms of spectral slopes; the coarser sample also reveals detectable absorption bands near 1.4, 1.9 and 2.2 μm , associated with OH/ H_2O (1.4 and 1.9 μm features) and aluminosilicates (2.2 μm feature)^[176].

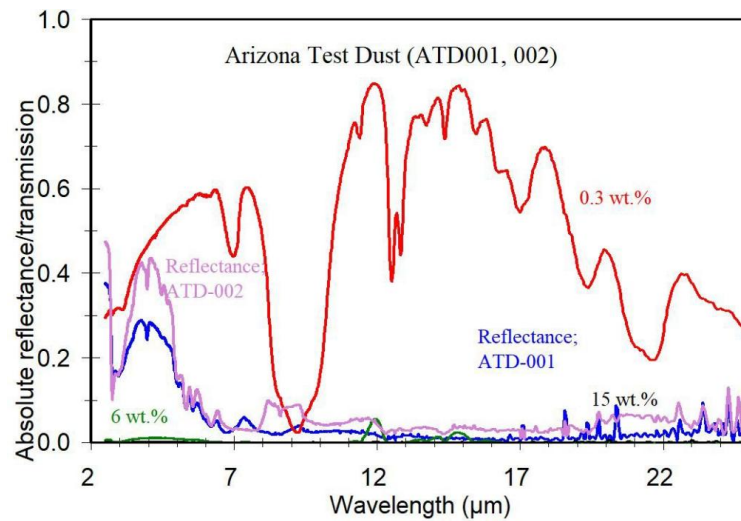


(a) 0.35–2.5 μm reflectance spectra of two grain sizes of Arizona Test Dust (<3 and <70 μm) and transmission spectra of the <3 μm sample for 0.3, 6, and 15 wt.% concentrations.

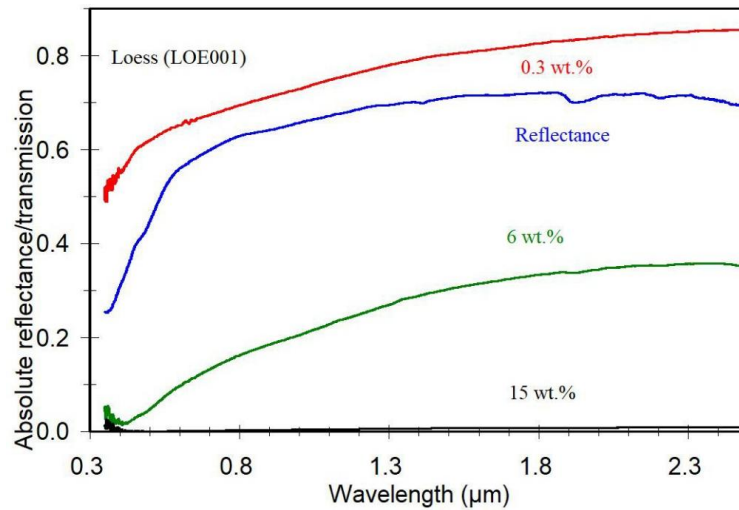


(b) 0.35–2.5 μm transmission spectra of Arizona Test Dust (ATD001) for the two highest concentrations.

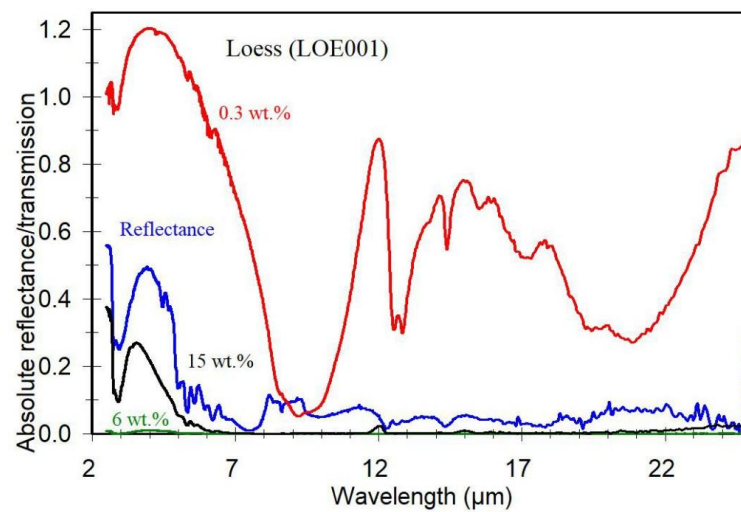
Figure 5. Cont.



(c) 2.5–25 μm reflectance spectra of two grain sizes of Arizona Test Dust (<3 and <70 μm) and transmission spectra of the <3 μm sample for 0.3, 6, and 15 wt.% concentrations.

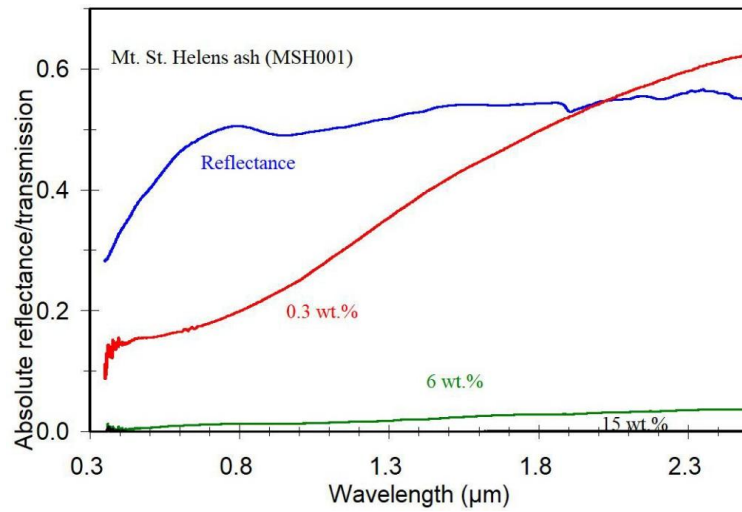


(d) 0.35–2.5 μm reflectance and transmission spectra of loess sample LOE001.

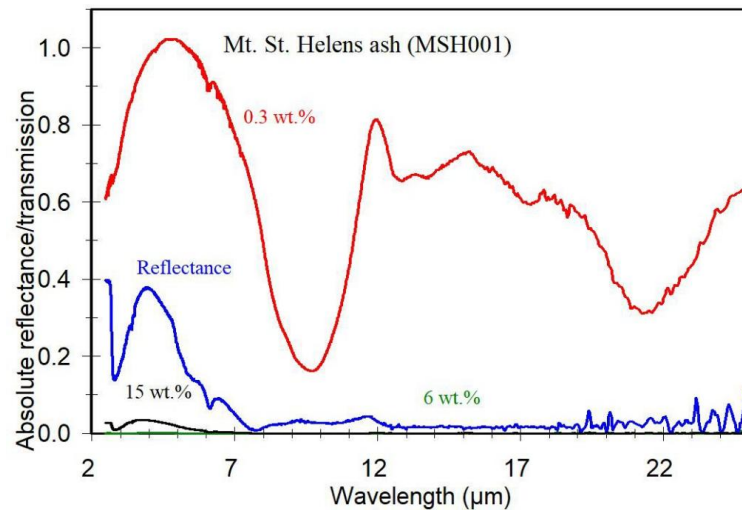


(e) 2.5–25 μm reflectance and transmission spectra of loess sample LOE001.

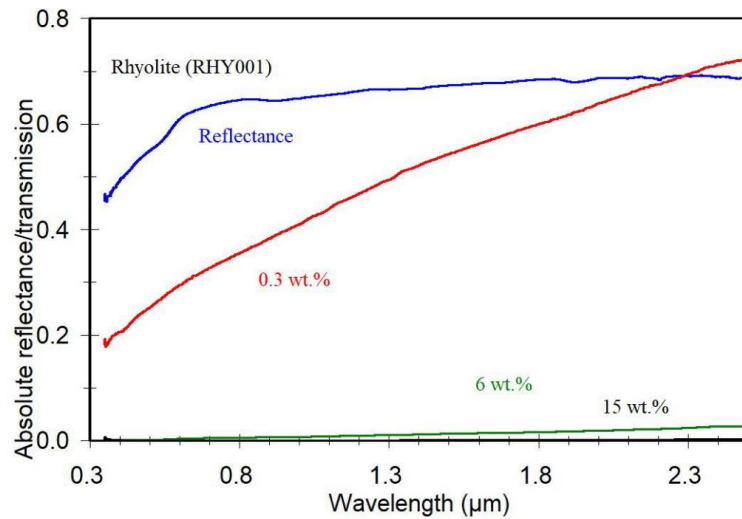
Figure 5. Cont.



(f) 0.35–2.5 μm reflectance and transmission spectra of Mt. St. Helens ash sample MSH001.

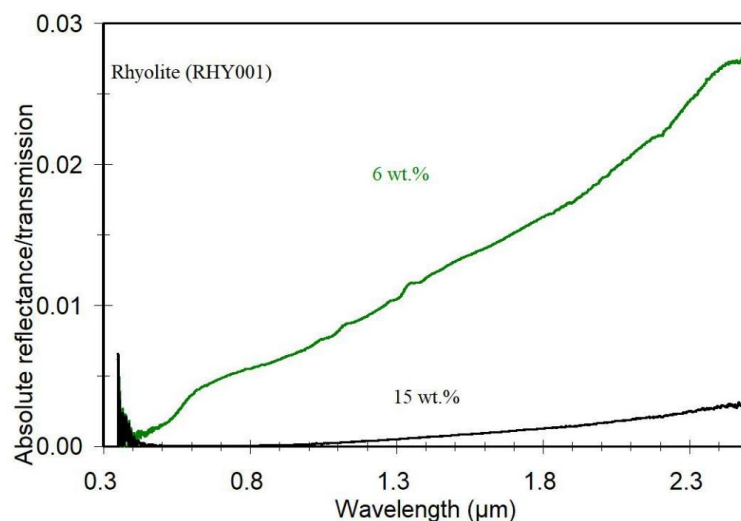


(g) 2.5–25 μm reflectance and transmission spectra of Mt. St. Helens ash sample MSH001.

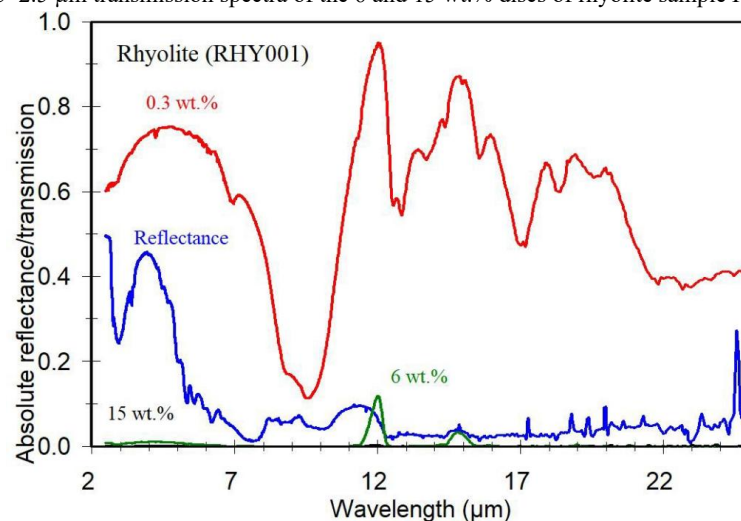


(h) 0.35–2.5 μm reflectance and transmission spectra of rhyolite sample RHY001.

Figure 5. Cont.



(i) 0.35–2.5 μm transmission spectra of the 6 and 15 wt.% discs of rhyolite sample RHY001.



(j) 2.5–25 μm reflectance and transmission spectra of rhyolite sample RHY001.

Figure 5. Rock/Mineral Mixture Group Spectra.

The KBr pellet spectra are red-sloped over the 0.35 to 2.5 μm region, in common with a number of the other silicate spectra (**Figure 5a**), with a 0.75/0.45 μm reflectance ratio of 1.51 for the 0.3% sample spectrum. At higher concentrations, transmission decreases dramatically, but an overall red spectral slope is preserved (**Figure 5b**). The 6 wt.% spectrum retains the best evidence of the 1.9 and 2.2 μm absorption features.

The 3–5 μm region is nearly featureless, with the exception of the large shoulder leading to the 3 μm water band and, in higher concentrations, the weak Si-O overtone feature at 3.95 μm (2530 cm^{-1}), seen best in the reflectance spectra (**Figure 5c**). The 8–13 μm window has the two distinct Si-O-(Si,Al) regions, with the asymmetric stretching region (950–1200 cm^{-1}) comprised of multiple overlapping absorp-

tion bands, and the corresponding symmetric region having the two bands at 12.50 and 12.82 μm (800 and 780 cm^{-1}). An additional absorption band is present at 11.36 μm (880 cm^{-1}) with is attributed to calcite^[188]. In terms of detail, it can be seen that the shape of the 8–10 μm absorption feature differs from that of the other silicates.

3.5.2. Loess (LOE-001) Spectra

Loess, deposits of windblown dust, may contain a wide range of materials, depending on source regions and transport^[223,224]. As with many of the silicate samples, the wind-blown silt sample (loess) provided little useful spectral information in the 0.4–2.5 μm (**Figure 5d**) and 3–5 μm windows (**Figure 5e**) with the exception of the weakly identifiable Si-OH absorption at 2.21 μm , large shoulders produced from

the water features outside of the 3–5 μm window, the weak Si-O overtones near 3.95 μm and weak organic features in the 3.33–3.57 μm (3000–2800 cm^{-1}) region. As with the other silicate-rich samples, it is red-sloped in the 0.35–2.5 μm region. XRD analysis showed the presence of quartz and albite.

The 8–13 μm window is, as with the other silicate-rich sample spectra, dominated by a very deep, broad feature, attributed to overlapping Si-O asymmetric stretching vibrations, with the corresponding symmetric stretches found in all of the silicate-bearing minerals in this study at 12.50 μm and 12.82 μm (800 and 780 cm^{-1}) (**Figure 5e**). Given the compositional makeup of this sample (**Table 2**) it is likely that these features undergo some level of Al for Si substitution.

3.5.3. Mt. St. Helens Ash (MSH-001) Spectra

The May 1980 eruption of Mt. St. Helens distributed ash over an extensive area^[225]. The erupted ash is silica rich, somewhat variable, but generally dacitic to rhyolitic in composition^[226] (**Table 2**). XRD indicates the presence of albite, quartz, and amorphous materials.

As with other silica-rich samples, it only exhibits weakly viable absorption features in the 0.4–2.5 μm window, most noticeably a broad, shallow feature around 1 μm attributable to iron, and a second weakly identifiable Si-OH feature around 2.21 μm ^[177] (**Figure 5f**).

Also similar to rhyolite (see below), the 3–5 μm window is largely spectrally featureless (**Figure 5g**), showing only the edges leading into H₂O features outside of the transmission windows. The 8–13 μm window is dominated by a very broad band, produced from overlapping asymmetric Si-O-(Si,Al) stretching features. It is more featureless than the same absorption in rhyolite, likely due to the ash having a greater amorphous/poorly crystalline component: the double absorption typically seen in other silicate bearing minerals appears to have lost fine details, consistent with more amorphous materials. The corresponding symmetric Si-O stretches are found at 12.58 and 12.82 μm (795 and 780 cm^{-1}).

3.5.4. Rhyolite (RHY001) Spectra

Rhyolite (a silica-rich igneous rock) was used in this study as a representative of explosive volcanic eruptions. It is expected to have spectral similarities to quartz, K-feldspar, and other Si-rich materials. As with the quartz sample, its 0.4–2.5 μm region spectrum is nearly featureless with the exception of a weak H₂O absorption band near 1.9 μm , a

weak Si-OH feature at 2.21 μm , and a probably Fe absorption band near 0.9 μm and reflectance dropoff towards the UV (**Figure 5h–i**).

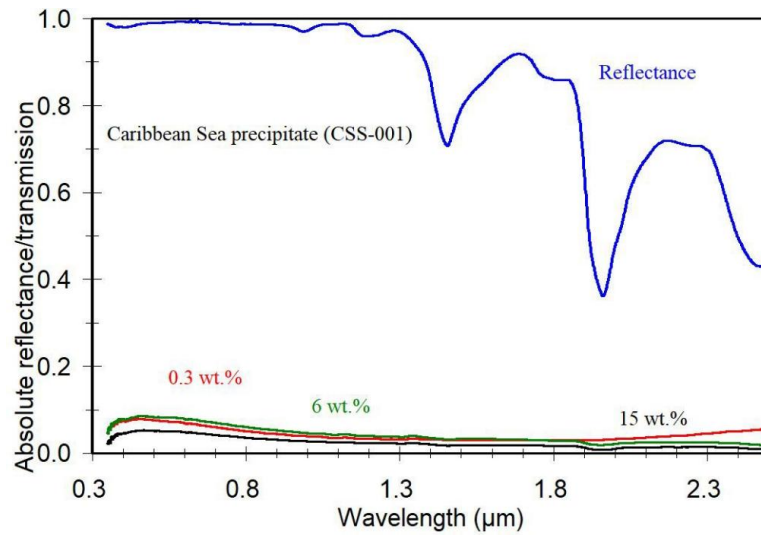
In the 3–5 μm window (**Figure 5j**), no strong absorption bands appear. In the 8–13 μm window the 0.3 wt.% spectrum shows a number of strong overlapping absorption features between 8.7 and 10 μm (1150 and 1000 cm^{-1}), arising from the asymmetric stretching of Si-O-(Si,Al). The corresponding symmetric stretches are seen at 12.50 μm and 12.82 μm (800 and 780 cm^{-1}). As expected, the rhyolite spectra are similar to the other silicates, such as quartz and K-feldspar, in showing strong Si and Al-associated absorption bands in the 9–10 μm region that are close to saturated in the 0.3 wt.% spectrum and saturated in the 15 wt.% spectra.

3.6. Ocean Water Precipitates Spectra

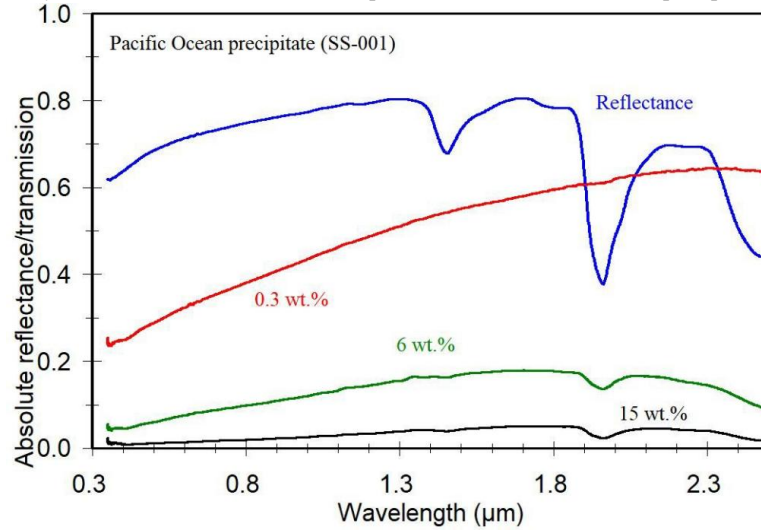
Sea spray is an important component of atmospheric aerosols^[227,228]. We used precipitates of seawater from two locations to understand its spectral properties (CSS-001 and SS-001). The two precipitates, produced by evaporation of seawater, show diverse spectral behaviour. The Caribbean Sea precipitate spectra (CSS-001; **Figure 6a**) show overall blue-sloped spectra over the 0.5–2.5 μm region, with a reflectance upturn in the 0.3 wt.% spectrum beyond ~2 μm . There is a transmission peak near 0.45 μm . There is little change in its spectral slope or transmission with changes in concentration.

Pure halite is expected to be spectrally featureless in the 0.35–2.5 μm region, and any absorption bands would be attributable to other phases. Water seems to be the major contributor to the spectra, with absorption features near 1.9 and 1.4 μm seen in all of the spectra. These lie within or at the edge of atmospheric water regions and are, therefore, unlikely to be of use in distinguishing aerosol sea salt. Given the high reflectance of halite, absorption featured due to other components in sea spray^[229,230] may appear in their spectra.

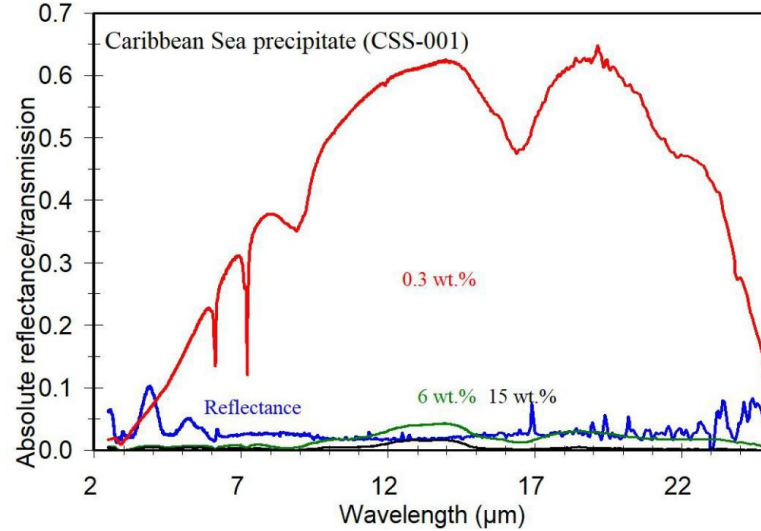
In contrast to the Caribbean precipitate, the Pacific precipitate transmission spectra show red slopes to ~1.7 μm (**Figure 6b**). As with the CSS-001 sample, the spectra are dominated by water-associated absorption bands, and the 1.95 μm band is evident in the higher concentration transmission spectra. Reflectance also varies systematically for these samples, with increasing concentration leading to lower overall transmission.



(a) 0.35–2.5 μm reflectance and transmission spectra of Caribbean Sea water precipitate CSS-001.

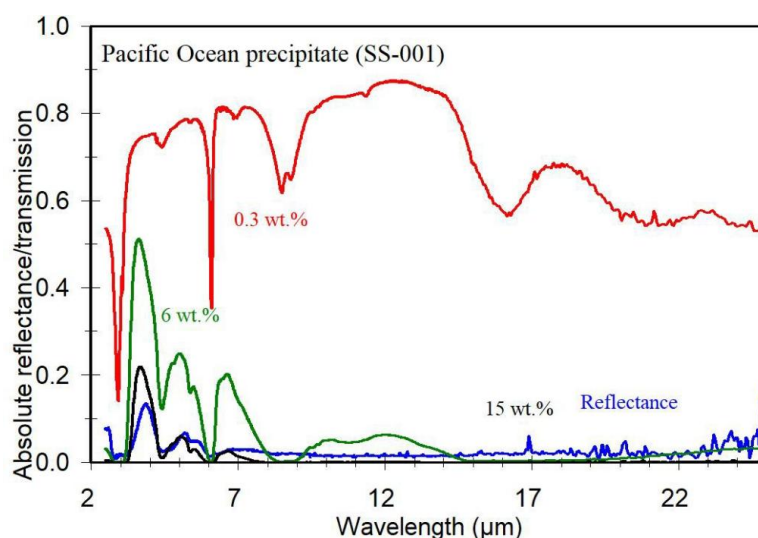


(b) 0.35–2.5 μm reflectance and transmission spectra of Pacific Ocean water precipitate SS-001.



(c) 2.5–25 μm reflectance and transmission spectra of Caribbean Sea water precipitate CSS-001.

Figure 6. Cont.



(d) 2.5–25 μm reflectance and transmission spectra of Pacific Ocean water precipitate CSS-001.

Figure 6. Ocean Water Precipitates Spectra.

In the 3–5 μm region both reflectance and transmission spectra have an absorption feature at 3.08 μm (3245 cm^{-1}) on the shoulder leading to the major water absorption, likely attributable to the first overtone of the H_2O fundamental bending vibration located in the 6 μm region^[176] (**Figure 6c,d**). Both samples show a major absorption feature around the 4.40 μm region (2275 cm^{-1}), however it is highly concentration-dependent, especially for the Caribbean sample which also shows the presence of a absorption feature at 4.64 μm (2155 cm^{-1}) at higher concentrations. The Pacific sample has an addition absorption feature around 4.82 μm (2075 cm^{-1}), which is missing from the Caribbean sample spectra. These features are likely attributable to the hydrated chloride (bischofite) present in the samples^[231] (**Table 3**).

The 8–13 μm window is expected to be a mix of numerous absorption features leading to complications in distinguishing individual features. Pure halite is spectrally featureless in this region and highly transparent, allowing small concentrations of other phases to contribute spectral features. The Pacific sample shows a broad double feature, with approximate bands centered at 8.47 and 8.77 μm (1180 and 1140 cm^{-1}). These features are absent in the 0.3% Caribbean sample, however they are strong at higher concentrations, with a slight shift in the second feature to 8.81 μm (1135 cm^{-1}). Short of 1000 cm^{-1} both samples have a broad absorption feature at higher concentrations; at 10.70 μm (935 cm^{-1}) for the Pacific sample and at 11.24 μm (890 cm^{-1}) for the Caribbean sample. These features are likely attributable

to the hydrated chlorides and calcite, as well as minor phases that may be undetectable by XRD.

It should be noted that sea spray is subject to a number of alteration processes in the atmosphere, so the spectra shown here are not definitive of sea spray aerosols or dust^[232].

4. Discussion

How to uniquely identify individual dust species from satellite observations is beyond the scope of this current paper; however, distinguishing between many key aerosol species in a laboratory setting appears to be feasible. While many individual samples show very similar spectral absorption features, mostly due to the presence of the same fundamental absorption groups, most of our dust species groups show potentially diagnostic absorption features in at least one of the defined atmospheric transmission windows.

4.1. Selected Characteristics and Materials of Interest

4.1.1. Urban Air Particulate Comparisons

The parking garage intake-filter particulate matter contains an array of absorption features from multiple species. While a range of dust species makes sense from a sample derived from a fan filter, it does lead to the conclusion that this sample is too variable for direct relevance in distinguish-

ing specific aerosol species without further sample treatment. However, it does provide a snapshot of urban dust, and future work will include direct measurement of all expected contributors in an attempt to derive the relative quantities of the various components. This could potentially lead to the ability to make inferences about the quantities of the individual species present.

4.1.2. Carbonaceous Aerosol Comparisons

The dominating C-H stretching vibrations in the 3.33–3.5 μm (3000–2850 cm^{-1}) region, shown in both the car exhaust and chimney ash, demonstrate the feasibility of distinguishing organic-rich aerosol species from mineral species. While we cannot directly distinguish between these two samples from this spectral region alone, the presence of carbonate minerals, remaining after ash combustion, is only present in the ash sample. Furthermore, it appears that there is a relative loss of organic absorption features in the ash spectra in the 8–13 μm window, which corresponds to the findings of a previous study^[233], which showed a strong reduction in organic absorption in this region with increased levels of combustion. This is not the case for the car exhaust particulate sample, as multiple overlapping organic vibrations still dominate. Possible aliphatic C-H overtone and combination absorption bands in the 2.3 μm region^[174] are not seen, likely due to a low abundance of aliphatic C-H molecules.

4.1.3. Silicate Mineral Comparisons

While it would be useful to be able to quantitatively determine the amounts of various mineralogical components present in, for example, a dust plume, it seems unlikely that this would be possible without a plume of extremely monomineralic composition. As previously mentioned, the two features at 12.50 and 12.82 μm (800 and 780 cm^{-1}) are indicative of silicate-bearing minerals, however, the large amount of overlap in the 8.33 and 11.11 μm (900 and 1200 cm^{-1}) range would make identification of individual species near impossible. Si-O stretching region of many tectosilicates shows broad undifferentiated absorptions and having access to the 12.5–25 μm (800–400 cm^{-1})^[184] region would be necessary for unique identification; however, this range mostly falls outside of the range of the atmospheric transmission windows. Nevertheless, a few indicators of mineral composition do exist. In both the kaolinite and illite

samples, the presence of OH deformation groups produces unique signatures in the 10.64–12.05 μm (940–830 cm^{-1}) region. In illite this creates a wider overall absorbance feature across the window, while in kaolinite, a distinct separate feature appears. Excluding these two samples and looking at the remaining silicate-bearing samples we see a general tendency for the band minimum in the asymmetric Si-O-(Si, Al) stretching region to shift to shorter frequencies with an increased Al: Al + Si ratio^[234]. This shift is also expected for the broader SCFM index ($\text{SiO}_2/(\text{SiO}_2 + \text{CaO} + \text{FeO} + \text{MgO})$)^[235,236]. This is somewhat confounded in our spectra, particularly with the potassium feldspar sample, as the overlapping absorptions and band saturation in this region do not produce a truly distinguishable minimum or minima. However, this broad undifferentiated absorption is expected of any samples with high orthoclase or intermediate plagioclase^[234]. Additionally, there is a general trend for the 12.5 μm (800 cm^{-1}) Si-O symmetric stretching absorption feature to increase in intensity with increasing SiO_2 content. This is in line with a previous study^[237] which examined 66 mixtures of quartz, orthoclase and oligoclase, finding that this band decreases in intensity with decreasing quartz content and that, in the cases where quartz content was constant, the band intensity would decrease with decreasing orthoclase or increasing oligoclase.

4.1.4. Mt. St. Helens Ash Properties

Distinguishing between our volcanic ash sample and those of a similar mineralogical makeup is somewhat difficult given the restricted atmospheric transmission windows. Upon visual comparison, the volcanic ash sample spectra are very similar to the other silicate-rich sample spectra, with the exception of the Si-O symmetric stretching region, which loses much of its double band structure, becoming a broader single feature. This feature, along with the Si-O symmetric bending vibration found outside of the atmospheric transmission window (695 cm^{-1}) has been used as a measure of crystallinity in SiO_2 rock samples^[238].

4.2. Utility of Atmospheric Windows

KBr is widely used in near-infrared studies as a medium for transmission measurements, however, its absolute transmission falls off sharply approaching the visible wavelength region^[239]. By using a blank KBr disc as the transmission

standard, spectral contributions from it are minimized, but not completely eliminated, as the quality and thickness of the pellets can vary from sample to sample. In addition, at the higher sample concentrations, the amount of KBr in the optical path will differ between the samples and the blanks. As a result, the general slope and absolute transmission in this region should be taken with a “grain of salt”. This also applies, mostly in terms of absolute transmission, in the mid-infrared regions. Below we discuss the spectroscopic properties identified for the major aerosol groups within each of the defined atmospheric windows.

Different spectral regions are sensitive to different aspects of dust composition, such as metal-oxygen charge transfers (visible region), crystal field transitions (visible-near infrared region), fundamental molecular vibrations (mid-infrared region), and their combinations and overtones (near-to mid-infrared region)^[240,241].

4.2.1. The 0.4–2.5 μm Window

Generally speaking, the 0.4–2.5 μm window showed low overall transmittance for many of the higher concentration sample transmission spectra, thereby potentially masking some absorption features. However, for the majority of samples, the 0.3 wt.% spectra were not saturated in any wavelength region. Many of the inorganic dust species produce excellent, identifiable absorption features in this region, although in some cases a higher concentration of sample would be beneficial to better identify some absorption bands (e.g., ammonium-bearing phase spectra – fundamental versus combination/overtone bands).

4.2.2. The 3–5 μm Window

Distinguishability of features relating to our broad categories of aerosols in the 3–5 μm is somewhat problematic, as individual samples from most groups produce similar features in a few key spectral regions within the window. Absorption features relating to organic materials (C-H stretching) can be prevalent in the 3.33–3.57 μm (3000–2800 cm^{-1}) region. Many of the samples presented here show very weak bands in this area, likely due to low abundances of aliphatic C-H groups, but a few contain strong bands in this region. Our carbonaceous aerosols can show strong absorption in this region, although contributions from carbonate overtones in the chimney ash sample somewhat distort the organic bands and certainly affect the depth of the features. Our am-

monium sulfate and nitrate sample spectra also show strong absorption in this region; however, the absorption features seen are the result of overlapping fundamentals, and, as such, produce a much broader absorbance feature. For the pure phases, these two classes are fairly easily distinguishable from each other. From our mineral and seawater precipitate categories, the 4–5 μm (2500–2000 cm^{-1}) regions are of interest. Overtones and combination bands of the fundamental SiO_4 and SO_4 absorptions are present in varying strengths in this region. These tend to produce multiple absorption features while in the seawater precipitate spectra a single, broad feature is present that is not seen in any other sample spectra.

4.2.3. The 8–13 μm Window

Due to the extensive overlapping of fundamental absorptions in this region, great care would be required in distinguishing between individual dust species. Almost every sample in this study shows at least one strong absorption in this region. That being said, a few potential indicator absorption features are present that may be useful in distinguishing between at least the different types of samples presented here. Carbonate-bearing samples all have a fairly narrow and deep absorption feature in the 10.81–12.0 μm (925–833 cm^{-1}) range owing to the out-of-plane bending vibration of CO_3^{2-} ^[188]. The silicate minerals/silicate-bearing sample spectra have generally strong absorbance features in the 8.3 to 11.1 μm (900 and 1200 cm^{-1}) range; however, the same can be said for the sulfate-bearing samples. Fortunately, the two symmetric Si-O stretches at 12.5 and 12.8 μm (800 and 780 cm^{-1}) are unique to the silicate-bearing samples we examined. The shape of the absorption features in this region, and position of individual absorption bands may be useful to distinguish individual species. Additionally, the position of the Si-O region of absorption may be useful to at least constrain the Si content of silicate-bearing dust^[235,236]. The shape of the 8–10 μm absorption feature differs between the silicate samples, but may not be diagnostic, as it reflects Si-O types of bonds, how many there are, and how amorphous the sample is.

4.3. Grain Size Effects

The CRB145 and ATD-001/002 samples allowed us to examine the effects of grain size on transmission spectra. Re-

flectance spectra of CRB145 showed a generally systematic decrease in reflectance across the entire examined spectra region with increasing grain size (**Figure 4a,d**). In transmission, systematic trends were not observed in the 0.35–2.5 μm spectra for either the 0.1 or 1 wt.% spectra (**Figure 4b,c**). This is likely due to incomplete mixing as we tried to minimize mechanical abrasion of the samples during mixing with KBr. The Fe^{2+} - and CO-associated absorption bands were also not confidently detected in any of the transmission spectra.

In the IR transmission windows, strong absorption bands are present and clearly seen in the reflectance spectra. Only a few of these bands, most notably the C-O band near 7.1 μm , were present in some of the 0.1 wt.% spectra and most of the 1 wt.% spectra (**Figure 4e,f**). Fortunately, from an observational perspective, the finest-grained sample spectra showed the strongest absorption bands.

4.4. Reflectance Versus Transmission

The main purpose of acquiring both reflectance and transmission measurements was to search for any spectral differences in terms of absorption band detectabilities. The reflectance spectra generally showed the most spectral detail, such as detectable absorption bands. Of the transmission spectra, the higher concentration samples (6 and 15 wt.%), showed more detail than the 0.3 wt.% spectra in the 0.35–2.5 μm region. Which spectrum was more useful in this regard depended on whether the absorption bands became saturated. In the infrared, the lowest concentration transmission spectra showed the greatest spectral contrast, allowing more absorption bands to be detected, as well as less saturation and hence loss of details in strong absorption bands (e.g., FAN-001, CHA-001).

4.5. Concentration Effects

The major purpose of measuring transmission spectra at three concentrations was to examine how concentration affects absorption bands of different intensities. In the 0.35–2.5 μm region, the 0.3 wt.% pellets generally showed the least amount of absorption bands and spectral contrast.

In the 3–5 and 8–13 μm regions, the 0.3 wt.% concentration spectra are adequate to show all the diagnostic absorption features; fundamental absorption bands are not

saturated. With increasing concentration, many of the diagnostic absorption bands saturate, and adjacent bands can merge into a region of low to no transmission. This makes spectral identification of phases more difficult, and as a result, many samples cannot be as precisely identified. As an example, diagnostic absorption bands in the infrared spectra of illite and kaolinite in the 9–13 μm region, blend into a single region of no transmission at 6 wt.%. As a result, some sort of silicate may be inferred, but a specific assignment would not be possible. As absorption bands become saturated, they widen, and hence wavelength positions of absorption band maxima cannot be as precisely determined. Adjacent absorption bands can also be subsumed into a broad region of low to no transmission.

4.6. Secondary Absorption Bands

Fundamental stretching and bending absorption bands, which largely occur in the 8–13 μm transmission window, can exhibit combinations and overtones in shorter wavelength regions, such as the 3–5 μm atmospheric window. For instance, illite's fundamental Si-O absorption bands occur in the 9–13 μm region, and are not saturated in the 0.3 wt.% spectrum. First order overtones are seen near 5 μm , but are weak in the 0.3 wt. % spectrum. They become more apparent in the 6 wt.% spectrum, where the fundamental bands are saturated, providing a means to detect illite at higher concentrations. A similar situation is seen, for example, in the SPT101 ammonium sulfate spectra, where weak overtone and combination bands are best seen in the 6 wt.% spectrum near 4.8 μm .

4.7. Implications for Aerosol Optical Depth (AOD)

Aerosol Optical Depth (AOD) represents the total aerosol load from the Earth's surface to the top of the atmosphere. The relationship between AOD and volume percent aerosols is complex and relies on several highly variable physical and optical properties of the aerosols^[102,115,242]. AOD is also wavelength dependent^[114,243,244]. For these reasons, deriving AOD from laboratory transmission spectra is highly uncertain and complex. Nevertheless, our laboratory spectra, covering a wide wavelength range, can provide valuable inputs to AOD modeling and determinations. Similarly,

our laboratory spectra can serve as useful inputs for derivation of optical constants^[245], from which detailed synthetic atmospheric transmission spectra can be derived^[246,247].

5. Conclusions

Laboratory transmission spectra can provide valuable insights into species that may be present in atmospheric dust, because dust composition is usually site-specific. Different types of aerosols/dust have unique spectral transmission and absorption properties that relate to their composition. Grain size variations mostly affect absorption/transmission intensities. Diagnostic spectral properties can exist in a number of atmospheric transmission windows, and diagnostic absorption bands can be used to identify dust species with varying degrees of specificity. In wavelength regions where absorption bands saturate, specificity is lost, but can be partially compensated for by the increasing depth of weaker absorption bands. Laboratory studies, of the kind carried out here, can provide experimental data that could be used to identify species present in the atmosphere and to help constrain measurements and models of atmospheric optical depth.

Supplementary Materials

The supporting information can be downloaded at <https://journals.bilpubgroup.com/files/JASR-12807-Supplementary-Materials.zip>.

Author Contributions

Conceptualization, E.C. and D.A.; methodology, C.H., M.C., P.M. and D.A.; software, D.A. and E.C.; validation, D.A.; formal analysis, C.H., S.M. and M.C.; investigation, C.H., M.C., P.M. and D.A.; resources, E.C.; data curation, D.A.; writing—original draft preparation, E.C., M.C. and C.H.; writing—review and editing, all; visualization, E.C.; supervision, E.C.; project administration, E.C. and D.M.; funding acquisition, E.C. All authors have read and agreed to the published version of the manuscript.

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Institutional Review Board Statement

This study did not require ethical approval.

Informed Consent Statement

Not applicable.

Data Availability Statement

Data associated with this study can be found on the lead author's laboratory website: <http://www.uwinnipeg.ca/c-tape>.

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Conflicts of Interest

The authors declare no conflict of interest.

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